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ANNUAL REPORT TO THE MANNED SPACECRAFT CENTER
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
For the Period Ending January 31, 1972

NUCLEAR CHEMISTRY OF RETURNED LUNAR SAMPLES:
NUCLIDE ANALYSIS BY GAMMA-RAY SPECTROMETRY

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CONTENTS

I. Introduction.	1
II. Completion of Measurements on Apollo 12 Samples . . .	2
III. Studies on Samples from the Apollo 14 Mission . . .	10
IV. K, Th, and U Concentrations and Cosmogenic Radio- nuclide Abundances in Rocks and Soils from Apollo 15: Preliminary Information	13
V. Recent Publications	15

NOTE

This is a report of work in progress. Many of the numerical data and suggested interpretations presented are preliminary or tentative. Those wishing to use or quote such information are requested to consult the authors regarding final results and status of publication.

I. INTRODUCTION

The period immediately following the Second Lunar Science Conference, January 11-14, 1971, was devoted to analyses of data on Apollo 12 samples which had not been completed earlier. In addition, samples of the upper portion of the double core (12025, 4-14) and rock 12031 were not received for analysis until March 11, rather later than expected. These tasks and the preparation of two papers on our Apollo 12 work (Section V) occupied the early part of the contract period.

The first sample received from the Apollo 14 mission at Oak Ridge National Laboratory (ORNL) was obtained in late May of 1971. Data on our entire collection of 11 samples were presented at the Third Lunar Science Conference, Houston, Texas, January 10-13, 1972 (see Section V).

The Apollo 15 mission provided a unique opportunity to laboratories such as ours, that are equipped to acquire and process data rapidly. Because no quarantine procedures were required, it was possible to measure concentrations of radionuclides with relatively short half-lives, i.e., half-lives of a few days or greater. This work was a natural extension of our assays of samples from Apollo 12 at short times after sample return.

Unlike previous missions, the Apollo 15 sample collection was not immediately preceded by a high-intensity solar flare; thus Apollo 15 presented a unique opportunity to test our earlier predictions of the galactic cosmic-ray production rates of short-lived radionuclides such as 16-day ^{48}V . In addition, our studies were expected to contribute other information such as concentrations of the primordial radioelements K, Th, and U, and long-lived cosmogenic radionuclides such as ^{26}Al and ^{22}Na .

A preliminary report of results on six samples from Apollo 15 was published in Science (see Section V). A more complete study of nine samples was presented at the Third Lunar Science Conference, Houston, Texas, January 10-13, 1972. Additional samples have been allocated and will be studied during the next reporting period.

These efforts will be described in more detail in the sections which follow.

II. COMPLETION OF MEASUREMENTS ON APOLLO 12 SAMPLES

J. S. Eldridge, K. J. Northcutt, G. D. O'Kelley

Concentrations of primordial radioelements and of cosmogenic radionuclides in crystalline rocks, breccias, and soils from the Ocean of Storms were determined both at the Lunar Receiving Laboratory (LRL) and at ORNL. Final data on samples which comprised about 40% of the Apollo 12 material were summarized in two papers (1, 2). Since these papers were submitted, rock 12031,2 has been analyzed for its major radioactive components; variations in the concentrations of ^{26}Al and ^{22}Na as a function of depth were determined for rocks 12002 and 12053; and studies on the Apollo 12 double core (12025, 12028) are nearing completion.

Results on 12031,2. - At the request of the Lunar Sample Analysis Planning Team (LSAPT), rock 12031,2 was measured non-destructively to determine whether any unusual radionuclide concentrations were present which would suggest a need for more extensive chemical analysis. The only difficulty encountered was the lack of a model of the rock. A suitable model was constructed with the aid of excellent documentary photography supplied by the LRL and direct observations of the rock through its clear plastic container. Radionuclide standards were prepared from this model in the same shape, electronic density, and bulk density as rock 12031,2. Preparation of the standards and the data analysis procedures were carried out as described previously (1-3).

The results obtained are:

Weight, 170.6 g
Density, 2.57 ± 0.13 g/cm³
K, 543 ± 20 ppm
Th, 0.86 ± 0.05 ppm (Th/U ~ 3.9)
U, 0.22 ± 0.02 ppm (K/U ~ 2470)
 ^{26}Al , 67 ± 5 dpm/kg
 ^{22}Na , 40 ± 5 dpm/kg
 ^{54}Mn , trace.

The concentrations of K, Th, and U are typical of an Apollo 12 mare basalt. The K/U ratio and the K concentration fit the systematics (2) compiled for other Apollo 12 samples. Concentrations of ^{26}Al and ^{22}Na suggest recent low exposure to solar cosmic ray bombardment, as if the rock were partially buried on the lunar surface just prior to collection. This point will be pursued with reference to lunar surface photography.

Concentration Variations with Depth in 12002, 12053.— Since each cosmogenic radionuclide has a characteristic energy threshold and excitation function for its production, variations in concentration with depth are to be expected, and can give important information concerning the energy distributions and fluxes of bombarding particles. By nondestructive gamma-ray spectrometry we determined approximate concentration gradients for ^{22}Na , ^{26}Al , ^{54}Mn , and ^{56}Co in samples (1,3) from Apollo 11 and Apollo 12. The high concentrations of these nuclides at rock surfaces arise from solar flare protons of low energy, which are effectively stopped in a few centimeters at most; on the other hand, galactic cosmic rays react throughout the volume of a rock weighing several kilograms.

We have applied the nondestructive gamma-ray spectrometry method to the study of concentration gradients in Apollo 12 rocks 12002 and 12053, whose selenographic orientations were known. From each rock was cut a set of samples forming a continuous series from top to bottom. Unfortunately, samples of 10-15 grams are needed to complete such measurements in a reasonable time; the samples we received were about 4-5 grams each. Thus, our accuracy is limited by counting statistics and by other uncertainties which arise when sample counting rates are near background rates.

The concentrations of ^{22}Na and ^{26}Al in these studies are presented in Table I and in Figs. 1 and 2. The results for 12002 are compared in Fig. 1 with measurements by Rancitelli et al. (4) on the same samples. Also shown in Fig. 1 are results of Finkel et al. (5), carried out by destructive techniques with excellent depth resolution, but shown averaged over approximately the same depth as in the present study. The points of Rancitelli et al. are shown at slightly different depths because of differences in measuring the dimensions of the rock slices and in the bulk densities used. The bulk densities noted in Table I were determined by us (1) in our studies of the whole rocks 12002 and 12053.

The error bars shown are due to counting statistics only. The data on rock 12002 in Fig. 1 show considerable scatter, but agree reasonably well. Our data show substantial deviations from the results of Finkel et al. (5), but agree more closely with data of Rancitelli et al. (4). The two sets of measurements on 12053 agree quite well, as shown in Fig. 2. Also shown in both illustrations is the galactic cosmic ray (GCR) production level for ^{26}Al and ^{22}Na in 12002 and 12053, calculated as we described previously (1).

Several models have been used to interpret radionuclide concentration variations with depth (4-6). The calculations of Armstrong and Alsmiller (6) utilize Monte Carlo techniques to obtain a detailed description of the induced nucleon-meson cascade at high energy. Spallation cross sections are computed using the intranuclear-cascade-evaporation model. Their calculated depth

Table I. CONCENTRATIONS OF ^{26}Al AND ^{22}Na AS A FUNCTION OF DEPTH
IN ROCKS 12002 AND 12053

Sample	Average Depth (g/cm ²) ^a	^{26}Al (dpm/kg)	^{22}Na (dpm/kg)
12002, 93	1.7	84 $\pm$ 5	62 $\pm$ 5
12002, 94	5.3	47 $\pm$ 5	31 $\pm$ 5
12002, 95	10.4	37 $\pm$ 3	27 $\pm$ 3
12002, 96	15.2	37 $\pm$ 4	26 $\pm$ 3
12002, 97	18.9	37 $\pm$ 5	26 $\pm$ 5
12053, 38	1.6	109 $\pm$ 5	54 $\pm$ 5
12053, 41	5.2	47 $\pm$ 5	35 $\pm$ 5
12053, 44 ^b	9.6	50 $\pm$ 5	27 $\pm$ 3
12053, 43	14.0	48 $\pm$ 4	23 $\pm$ 5
12053, 42	18.3	38 $\pm$ 4	26 $\pm$ 5
12053, T ^c	10.3	55 $\pm$ 4	31 $\pm$ 3

^a Measured density of 12002 = 3.14 g/cm³; for 12053 = 3.49 g/cm³.

^b Average of 12053, 41, 44, 43.

^c Average of all 5 slices: 12053, 38, 41, 42, 43, 44.

Average concentrations of Th = 0.80 $\pm$ 0.04; U = 0.27 $\pm$ 0.03.

ORNL-DWG. 71-9998

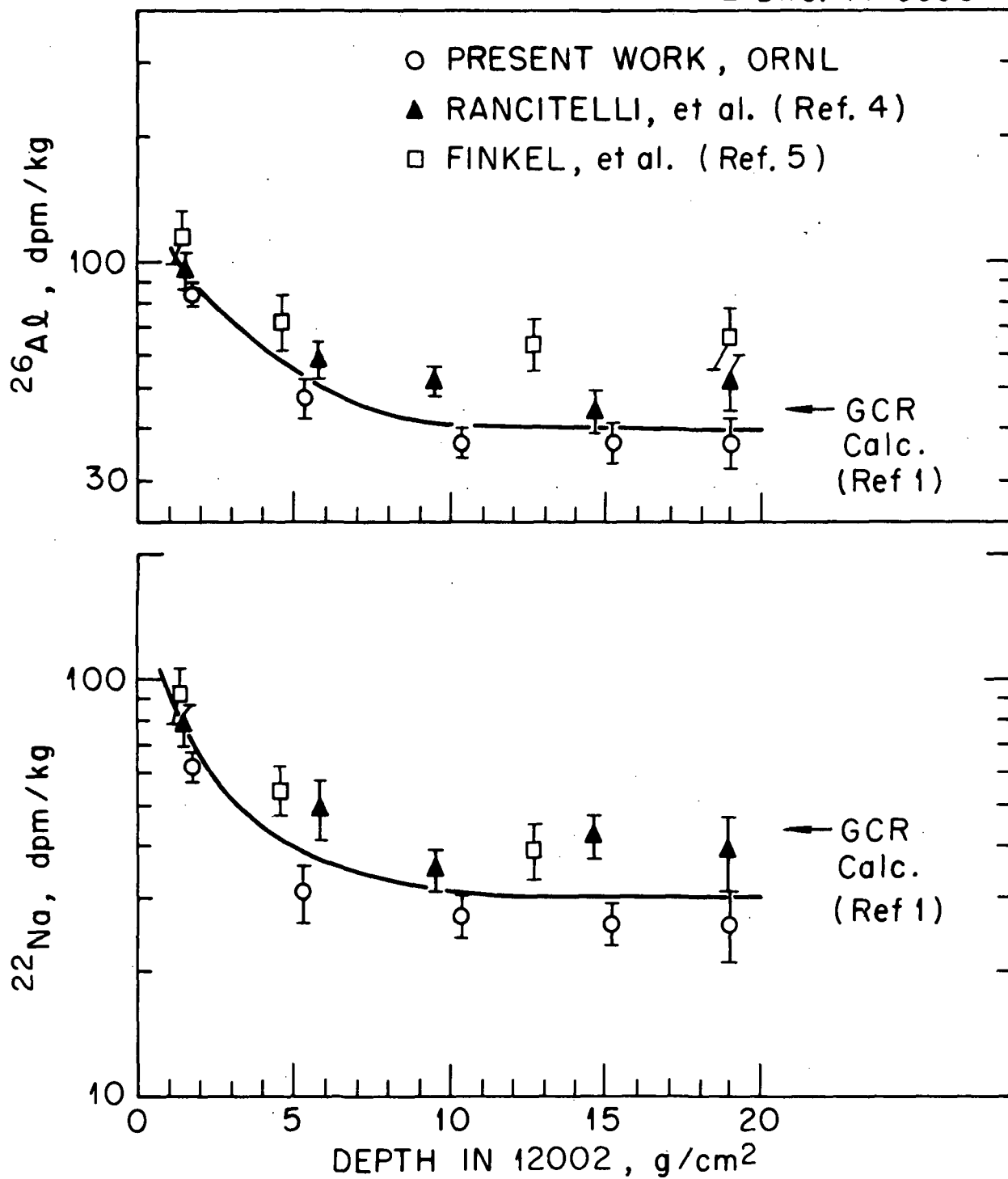


Figure 1. Variations in concentrations of ^{26}Al and ^{22}Na with depth for rock 12002.

ORNL-DWG. 71-9999

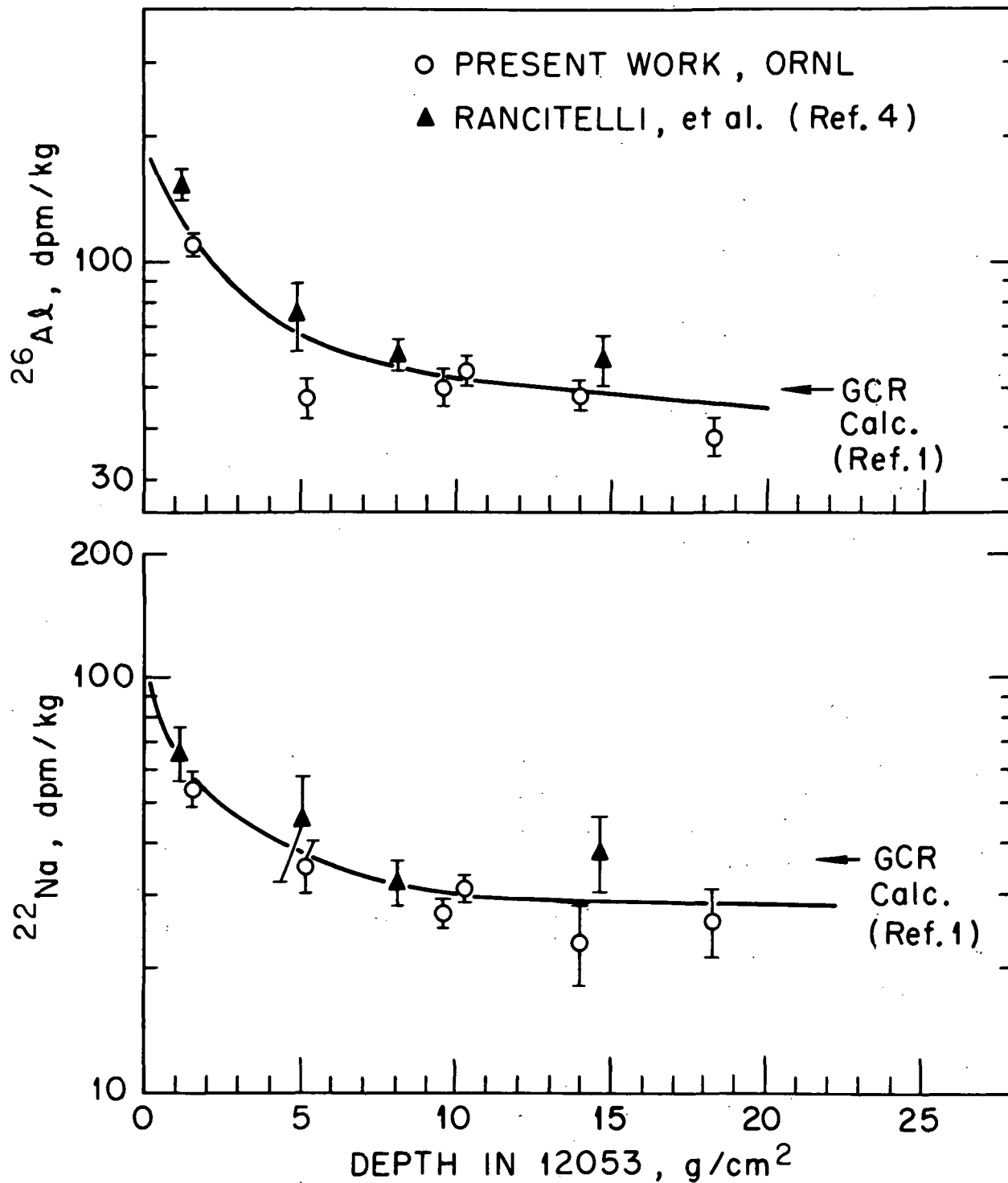


Figure 2. Variations in concentrations of ^{26}Al and ^{22}Na with depth for rock 12053.

dependence (6) of ^{26}Al and ^{22}Na is in good agreement with our results of Table I, although their calculation was based on an average lunar sample composition, which differed in detail from the actual chemical compositions of our samples. In the new contract period we hope to carry out further calculations with the Armstrong and Alsmiller program in terms of the known chemical concentrations and densities of 12002 and 12053. We hope to evaluate the degree to which this model may be used to deduce bombarding particle fluxes and energy distributions from depth variations.

Concentrations of ^{26}Al and ^{22}Na in the Double Core 12025, 12028.— Measurement of the double core from Apollo 12 has proved very difficult because of the small samples involved. The top five samples (12025, 10, 11, 12, 13, 14) range from only 0.75 gram for 12025, 14 to about 1.7 grams for 12025, 11. Samples deeper in the core are 1 to 2 grams each. From about 3 cm (5.4 g/cm^2) down, it was necessary to measure samples in groups to compensate for the decrease in radioactive concentrations with depth.

The results to date are shown in Table II. Because of the low radioactivity of such small samples and limited counting time, statistical accuracy is poor. In addition, a better background spectrum is required for the small samples 12025, 10-14. Shortly we hope to improve the quality of the data in Table II with a more refined computer least squares analysis. The data in Table II generally agree within error limits with similar measurements by Rancitelli et al. (4).

The rather complete pattern reported here for concentrations of K, Th, and U in the core samples points out the pronounced chemical stratification which may occur. This effect was first reported by Schnetzler and Philpotts (7), who found that alkali metal concentrations such as Li, K, and Rb were 3-10 times lower in 12028, 16 than in other regions of the Apollo 12 core. In their visual examination of the core, the Lunar Sample Preliminary Examination Team (LSPET) noted (8) a coarse-grained layer of glass, olivine crystals, and gabbro fragments at 13.2-15.3 cm. Core samples 12028, 16, 17 almost exactly bracket this layer. Our chemical data on K, Th, and U in Table II show decreases in concentration of about a factor of two for the "six-pack" of core samples 12028, 11-18. Our measurements of K concentration are consistent with a weighted average of 280 ppm K in 12028, 16, 17 and an average K concentration of about 2600 ppm for the other four of the group. Thus, it appears that the coarse layer is responsible for the abrupt reduction in chemical concentrations.

The existence of such concentration variations emphasizes the need for chemical analysis of all core tube material scanned by radioactivity methods, since the interpretation could be seriously compromised by unexpected fluctuations in abundances of the target elements responsible for the nuclides in question.

Table II. CONCENTRATIONS OF ^{26}Al , ^{22}Na , K, Th, AND U IN THE DOUBLE CORE 12025, 12028

Sample(s)	Average Depth (g/cm ²) ^a	^{26}Al (dpm/kg)	^{22}Na (dpm/kg)	K (ppm)	Th (ppm)	U (ppm)
12025, 14	0.4	196 ± 50	355 ± 105		5.9 ± 1.3	
12025, 13,12	1.8	165 ± 20		2700 ± 1500	5.3 ± 0.6	
12025, 11	3.8	50 ± 25	150 ± 70		7.0 ± 1.2	
12025, 10	5.2	40 ± 30	160 ± 70		6.6 ± 1.0	
12025, 9,8,7	8.5	48 ± 8	47 ± 17	2200 ± 300	6.1 ± 0.4	
12025, 6,5,4	13.5	50 ± 10	-	2300 ± 300	5.9 ± 0.2	
12028, 11-18	23.8	-	25 ± 5	1350 ± 100	2.9 ± 0.1	0.80 ± 0.04
12028, 20-26	35.5	-	20 ± 9	2950 ± 100	6.8 ± 0.2	2.0 ± 0.2
12028, 27-36	49.2	-	29 ± 6	3050 ± 200	7.2 ± 0.4	2.2 ± 0.2
12028, 37-44	64.3	-	-	2910 ± 200	6.5 ± 0.2	1.8 ± 0.2

^aBased on measured density of 1.81 for 12070 (Ref. 1).

REFERENCES

1. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Cosmogenic Radionuclide Concentrations and Exposure Ages of Lunar Samples from Apollo 12," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1747-1755, MIT Press, 1971.
2. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Abundances of the Primordial Radionuclides K, Th, and U in Apollo 12 Lunar Samples by Nondestructive Gamma-Ray Spectrometry: Implications for Origin of Lunar Soils," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1159-1168, MIT Press, 1971.
3. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Primordial Radionuclide Abundances, Solar-Proton and Cosmic-Ray Effects, and Ages of Apollo 11 Lunar Samples by Non-Destructive Gamma Ray Spectrometry," Proceedings of the Apollo 11 Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 1, Vol. 2, pp. 1407-1423, Pergamon, 1970.
4. L. A. Rancitelli, R. W. Perkins, W. D. Felix, and N. A. Wogman, "Erosion and Mixing of the Lunar Surface from Cosmogenic and Primordial Radionuclide Measurements in Apollo 12 Lunar Samples," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1757-1772, MIT Press, 1971.
5. R. C. Finkel, J. R. Arnold, M. Imamura, R. C. Reedy, J. S. Fructer, H. H. Loosli, J. C. Evans, and A. C. Delany, "Depth Variation of Cosmogenic Nuclides in a Lunar Surface Rock and Lunar Soil," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1773-1789, MIT Press, 1971.
6. T. W. Armstrong and R. G. Alsmiller, Jr., "Calculation of Cosmogenic Radionuclides in the Moon and Comparison with Apollo Measurements," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1729-1745, MIT Press, 1971.
7. C. C. Schnetzler and J. A. Philpotts, "Alkali, Alkaline Earth, and Rare-Earth Element Concentrations in some Apollo 12 Soils, Rocks, and Separated Phases," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, Vol. 2, pp. 1101-1122, MIT Press, 1971.
8. Lunar Sample Preliminary Examination Team, "Preliminary Examination of Lunar Samples from Apollo 12," Science, 167, 1325-1339 (1970).

III. STUDIES ON SAMPLES FROM THE APOLLO 14 MISSION

J. S. Eldridge, K. J. Northcutt, G. D. O'Kelley

The distribution schedule for Apollo 14 samples precluded any studies of short-lived radionuclides in our laboratory. In fact, the distribution of Apollo 14 samples was delayed such that about half of our samples were not received until July and August. Thus, we found that our measurements on Apollo 14 samples seriously overlapped our studies of Apollo 15 samples.

Concentrations of K, Th, U, ^{26}Al , and ^{22}Na were determined for seven clastic or brecciated rocks, three sieved samples of fines, and one composite sample of sawdust from extensive cutting of a fragmental rock. Two features of the data are apparent from a cursory examination: (1) levels of the primordial radionuclides are higher than those of the Apollo 11 and 12 samples by as much as a factor of ten; and (2) the spread in the concentrations of these radioelements is very narrow in all samples listed. A detailed report of our study was presented at the Third Lunar Science Conference (see Section V).

The material returned from Fra Mauro Base sampled a wide area containing Cone crater ejecta from depths > 10 m, as well as surface materials with long exposure ages. From an analysis of our sample suite coupled with those of KEITH et al. (1) we can calculate an average thorium content of 12.9 ppm for 24 samples weighing 29 kg out of the total sample inventory of 43 kg. Thus, at least 68 per cent by weight of the entire Fra Mauro sample collection contains ~ 13 ppm Th, and from the Th/U ratio of ~ 3.6 and the K/U ratio of ~ 1400 , we calculate an average uranium content of ~ 3.6 ppm and an average potassium content of ~ 5000 ppm. To the extent that the Fra Mauro sample collection is considered a representative sample of the Fra Mauro formation and Imbrium Basin ejecta, we can speculate that these concentrations of K, Th, and U will be found in a wide area of the lunar surface in and around the Imbrium Basin.

A simple two-component mixing diagram fitting our K/U systematics developed for Apollo 12 soils and breccias (2) indicates ~ 60 to 85 per cent content of foreign component (KREEP) in our collection of Apollo 14 soils and breccias. This finding is in excellent agreement with that of SCHONFELD (3), who used a 26-element linear mixing model and found a 60 to 95 per cent KREEP content in a larger group of Apollo 14 soils and breccias.

Sample 14321,38 is an 1100-g piece sawn from the north side of the 8996-g 14321 collected near station C1 during the second EVA. The lunar orientation was well documented by lunar surface photographs. Cutting diagrams prepared by the NASA-MSD Curator's

staff indicate that section 14321,38 has a top surface area of $\sim 60 \text{ cm}^2$ from a total top surface area of $\sim 420 \text{ cm}^2$. Sample 14321,256 is a 200-g aliquot of $\sim 1 \text{ kg}$ of sawdust obtained from multiple slab-and-wire-saw cuttings of rock 14321. WRIGLEY (4) measured Th, U, ^{26}Al , and ^{22}Na concentrations in 14321 sawdust and obtained excellent agreement with values shown in Table 1. KEITH (1) measured radionuclide concentrations in 14321,38 and obtained good agreement with our values.

Sawdust analyses should be quite valuable for over-all whole rock compositions for complex materials such as the Apollo 14 clastic rocks if suitable corrections are made for dilutions by the saw-wire debris. From a comparison of our 14321 sawdust and 14321,38 rock values, we can calculate an average dilution of the sawdust of ~ 15 per cent based on the dilution of the thorium content from the rock. Potassium and uranium dilutions yield different decrements, but the approximate 15 per cent value is within the error range of the potassium and uranium dilutions. RHODES (5) found contamination of ~ 15 per cent copper along with an unknown contamination of a fibrous material in the sawdust from the cutting of the breccia 14307. It is obvious that sawdust samples can be used for a variety of determinations to obtain whole rock or average concentrations for heterogeneous materials; however, the homogenized sawdust should be carefully analyzed in order to determine the type and quantity of diluents added by the cutting process.

The distribution schedule for Apollo 14 samples precluded any studies of short-lived radionuclides in our laboratory, since 115 days had elapsed after the samples left the moon and 127 days after the intense solar flare of January 25, 1971, before we received our first sample (14321,38). In addition, the high concentrations of thorium and uranium tended to mask the minor cosmogenic radionuclides. Concentrations of ^{26}Al and ^{22}Na were determined in all samples (Section V). Cosmogenic radionuclide determinations reported here show little differences from those found in previous Apollo missions with the exception of the three soil samples, which show unexpected results.

The Soil Mechanics Experiment trench was planned to be a 60 cm-deep trench about one crater diameter away from North Triplet crater at station G. The astronauts were instructed to dig the trench with one vertical sidewall to provide a means for sampling at depth. The trenching did not yield a vertical side wall; sloping occurred with walls of 60° - 80° , and a maximum depth of 36 cm was achieved. Samples 14148, 14149, and 14156 in our study were taken from the top, bottom, and middle, respectively, of the trench and are all $< 1 \text{ mm}$ sieved fractions. It was expected that there would be pronounced decreases in the concentrations of the cosmogenic species ^{26}Al and ^{22}Na with depth. Instead, all three samples show a surprising uniformity in concentrations of these nuclides. Due to the uniform distribution of ^{26}Al and ^{22}Na and their high concentrations in the "bottom" sample, we must conclude

that extensive mixing occurred, and sample 14149,62 is not representative of the soil at a 36-cm sampling depth. This also gives reason to question the apparent uniformity of K, Th, and U concentrations in the different soil layers. In addition, the separation of the < 1 mm fraction from the trench bottom sample has further emphasized the sampling defect, because the bottom sample has a median grain size of 0.41 mm compared to 0.09 and 0.007 mm for the surface and middle trench samples (6). Sample 14140, the 4-10 mm sieved fraction, is probably a more representative sample to characterize the trench bottom.

REFERENCES

1. J. E. Keith, R. S. Clark, and K. A. Richardson, "Gamma Ray Measurements of Apollo 12, 14, and 15 Lunar Samples (abstract)." In Lunar Science-III (editor C. Watkins), pp. 445-448, Lunar Science Institute Contr. No. 88, 1972.
2. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Abundances of the Primordial Radionuclides K, Th, and U in Apollo 12 Lunar Samples by Nondestructive Gamma-Ray Spectrometry: Implications for Origin of Lunar Soils," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta., Suppl. 2, Vol. 2, pp. 1159-1168, MIT Press, 1971.
3. E. Schonfeld, "Component Abundances and Ages in Soils and Breccia (abstract)." In Lunar Science-III (editor C. Watkins), pp. 683-685, Lunar Science Institute Contr. No. 88, 1972.
4. R. C. Wrigley, "Radionuclides at Fra Mauro (abstract)." In Lunar Science-III (editor C. Watkins), pp. 814-815, Lunar Science Institute Contr. No. 88, 1972.
5. M. Rhodes, private communication of unpublished results, NASA-MSC.
6. Lunar Sample Preliminary Examination Team, "Preliminary Examination of Lunar Samples from Apollo 14," Science, 173 681-693 (1971).

IV. K, Th, AND U CONCENTRATIONS AND COSMOGENIC RADIONUCLIDE ABUNDANCES IN ROCKS AND SOILS FROM APOLLO 15

G. D. O'Kelley, J. S. Eldridge,
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The Apollo 15 landing marked a notable scientific advance over previous Apollo missions. The increased astronaut mobility made possible the exploration of a large and diverse area of the Hadley-Apennine region. Well-documented rocks and rock fragments with a wide range of textures were returned, together with a variety of soil samples with significant geochemical differences from the igneous rocks and from each other. Thus, the samples from the selenological structures of the region offer unique opportunities to study the detailed geochemistry of the Apollo 15 landing site.

The techniques of nondestructive gamma-ray spectrometry have proved to be very useful for scanning a large number of samples to determine the concentrations of K, Th, and U. In addition to such chemical data, it is also possible to study in some detail the irradiation history of lunar samples by measuring the concentrations of radionuclides produced in the bombardment of lunar surface material by the solar and galactic cosmic rays. A preliminary account of measurements on some of the Apollo 15 samples reported below was published by O'KELLEY et al. (cf., reprint in Section V).

Samples from the Apollo 15 manned lunar landing have special significance to nuclear geochemistry because, unlike previous missions, sampling was not closely preceded by an intense solar flare. Thus, the Apollo 15 materials could be used to determine the galactic production rates of some short-lived radionuclides which previously were detected chiefly as products of solar-proton bombardment. Because no quarantine restrictions were imposed on samples from Apollo 15, it was possible to obtain samples for time-dependent studies in our laboratory rather soon after their arrival at the Lunar Receiving Laboratory (LRL).

Upon a recommendation by LSAPT and approval by NASA Headquarters, we were able to receive samples early in the Apollo 15 mission. Two samples were received from the LRL on August 11, 1971, about nine days after lift-off from the moon. Two weeks later another pair of samples was received. By the end of the present reporting period, a total of nine samples had been received and analyzed. Results of these studies were presented at the Third Lunar Science Conference, Houston, Texas, January 10-13, 1972 (see Section V).

Samples from Hadley Base which we investigated were basalts 15016, 15475, and 15495; in breccias 15285 and 15455; and in soils 15031, 15041, 15101, and 15601. The basalts of Apollo 15 are somewhat lower in K, Th, and U (respectively about 400, 0.54, and 0.14 ppm) than basalts of Apollo 12 or the Low-K basalts of Apollo 11 (about 520, 0.97, and 0.25 ppm). KREEP is ubiquitous in the Apollo 15 soils and breccias and ranges from 8 to 21% in the samples studied. Samples of soil or soil breccia from the mature, relatively undisturbed mare regolith and from the Apennine Front are highest in KREEP, while a sample of soil from the edge of Hadley Rille was significantly lower.

Two rocks have concentrations of ^{26}Al less than their saturation values; 15475 and 15495 may have been ejected onto the lunar surface as recently as 0.7 million years and 2.0 million years, respectively. Trench samples from the LM-ALSEP site show variations in radionuclide concentrations with depth similar to those of the Apollo 12 cores. The galactic cosmic-ray production rate of ^{48}V was determined as 57 ± 11 dpm/kg Fe. The concentration of ^{56}Co in rock 15016 leads to the conclusion that the solar flare of 24 January 1971 was 1.91 ± 0.45 times more intense than the flare of 3 November 1969, in agreement with other radiochemical data and with preliminary satellite measurements.

Further information will be found in the publications of Section V below. Additional samples have been allocated and will be analyzed in the next reporting period. These additional samples are: crystalline rocks 15058, 15065, 15076, 15379, 15476, 15486, 15499, 15545, 15557, and 15597; breccia 15498; and soil 15471.

V. RECENT PUBLICATIONS (ATTACHED)

- A. Abundances of the Primordial Radionuclides K, Th, and U in Apollo 12 Lunar Samples by Nondestructive Gamma-Ray Spectrometry: Implications for Origin of Lunar Soils.
- B. Cosmogenic Radionuclide Concentrations and Exposure Ages of Lunar Samples from Apollo 12.
- C. Primordial Radioelements and Cosmogenic Radionuclides in Lunar Samples from Apollo 15.
- D. Abundances of Primordial and Cosmogenic Radionuclides in Apollo 14 Rocks and Fines (abstract).
- E. Concentrations of Primordial Radioelements and Cosmogenic Radionuclides in Apollo 15 Samples by Nondestructive Gamma-Ray Spectrometry (abstract).

Cosmogenic radionuclide concentrations and exposure ages of lunar samples from Apollo 12

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Abstract—Cosmogenic radionuclide abundances in a suite of samples from the Ocean of Storms were determined nondestructively by gamma-ray spectrometers of low background. Samples investigated were crystalline rocks 12002, 12004, 12039, 12052, 12053, 12054, 12062, and 12064; breccias 12013, 12034, and 12073; fines 12032 and 12070. The general concentration patterns of spallogenic radionuclides resemble those observed for Apollo 11 samples, but with some differences in detail. Cosmogenic radionuclides determined in this study were ^{22}Na , ^{26}Al , ^{46}Sc , ^{48}V , ^{52}Mn , ^{54}Mn , ^{56}Co , and ^{60}Co . Despite delays in obtaining samples during the preliminary examination, 5.7-day ^{52}Mn was determined in two rocks and 16-day ^{48}V was determined in four rocks.

Solar protons and galactic protons are both involved in the production of ^{22}Na , ^{26}Al , and ^{54}Mn in surface samples; however, several rocks show evidence of shielding. Concentrations of radionuclides in rock 12034 are consistent with production by galactic protons at depth, shielded from the effects of solar protons. Sample 12002,30 from the top of rock 12002 exhibited high concentrations of nuclides produced by solar flare protons, in confirmation of the orientation of 12002.

From the ^{60}Co concentration in rock 12002, a thermal neutron flux of 0.35 ± 0.18 neutrons $\text{cm}^{-2} \text{sec}^{-1}$ was estimated. Estimates of cosmic-ray exposure ages were calculated by the ^{22}Na – ^{22}Ne method. The results for seven samples are in good agreement with ^3He exposure ages by other investigators and range from 48 to 251 million years.

INTRODUCTION

THE EXTENSIVE studies of nuclides produced in the bombardment of meteorites by the solar and galactic cosmic rays have revealed much detailed information concerning the intensity and energy spectra of the incident radiations and their constancy with time. Lunar samples are even more suitable objects for such studies than meteorites, since the lunar samples have been irradiated in known orientations in space and are free from atmospheric ablation. A number of studies on radionuclide concentrations in Apollo 11 lunar samples (BEGEMANN *et al.*, 1970; HERZOG and HERMAN, 1970; O'KELLEY *et al.*, 1970a, 1970b; PERKINS *et al.*, 1970; SHEDLOVSKY *et al.*, 1970; WRIGLEY and QUAIDE, 1970) clearly demonstrated the potential of such information for elucidating the bombardment history of the lunar material, the histories of the incident particle fluxes, the erosion rates of rocks, and the rate of turnover of the lunar surface due to impact.

The absence of atmospheric ablation makes possible the detailed study of the effects of recent solar flares and long-term solar particle bombardment. Because of

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the short range of the solar particles and the high yields of some of the nuclear reaction products, gamma-ray spectrometry has been used quite successfully to determine the most recent orientation of rocks on the lunar surface (O'KELLY *et al.*, 1970a, 1970b; PERKINS *et al.*, 1970; SCHONFELD and O'KELLY, 1971).

Several radionuclides of interest have short half-lives. For this reason, much of the data reported below were recorded at the Lunar Receiving Laboratory (LRL), Houston, Texas, during the preliminary examination of the Apollo 12 samples. An early account of the results on some of the samples was given in LSPET (1970). Since the publication of the preliminary examination report the data analyses have been refined and further samples have been analyzed.

EXPERIMENTAL PROCEDURES

Several gamma-ray spectrometers were used in the course of this study. A NaI(Tl) scintillation coincidence spectrometer with an associated on-line, data acquisition system described by O'KELLY *et al.* (1970b), together with a Ge(Li) spectrometer permitted rapid analyses of samples at the LRL during the quarantine period, so that nuclides of short half-life could be determined. Some studies at later times were carried out at Oak Ridge National Laboratory on a NaI(Tl) spectrometer similar to the scintillation spectrometer at the LRL.

The first Apollo 12 sample for radioactivity determination (12002,0) was received from the LRL Sample Laboratory on November 28, 1969, about 8.4 days after liftoff from the moon. During quarantine, samples were mounted in stainless steel containers for gamma-ray analysis. After quarantine, samples were generally sealed inside thin teflon bags for measurement. Methods of data acquisition and data analysis were essentially the same as those we used to analyze Apollo 11 samples.

For analyses of data on samples measured during the preliminary study, calibration of the LRL coincidence spectrometer was established by recording a library of spectra from cylindrical radioactive standards prepared by dispersing known amounts of radioactivity in quantities of iron powder. When recording the library of standard spectra, the standard sources were placed inside the steel containers actually used.

Spectrum libraries used for analyzing samples 12002,0; 12002,20; 12013,11; 12032,16; 12034,0; 12070,0; and 12073,0 were obtained from replicas which accurately reproduced the electronic and bulk densities of the lunar samples. Procedures for preparation of the cylindrical standards and the replicas were described earlier by O'KELLY *et al.* (1970a, 1970b). A more detailed description of the analytical procedures employed for the Apollo 12 studies was given by O'KELLY *et al.* (1971).

RESULTS AND DISCUSSION

Cosmogenic radionuclide concentrations

Our results on spallogenic radionuclides are given in Table 1. The general concentration patterns resemble those we observed in the Apollo 11 samples (O'KELLY *et al.*, 1970a, 1970b); however, a number of subtle differences were noted due to effects of chemical composition and shielding. The data of Table 1 were recorded on large samples, usually a rock or a large fragment of a rock. Sample weights are listed in a companion paper by O'KELLY *et al.* (1971). As observed in all gamma-ray spectrometry studies of Apollo 11 samples, the high concentrations of Th and U in lunar material makes difficult the determination of weak gamma-ray components. Because of the short times available for some of the measurements, it was not possible

Table 1. Concentrations (dpm/kg) of spallogenic radionuclides in Apollo 12 samples.* Values for short-lived nuclides have been corrected to 1426 GMT, Nov. 20, 1969.

Sample†	Type‡	²² Na	²⁶ Al	⁴⁸ Sc	⁴⁸ V	⁵² Mn	⁵⁴ Mn	⁵⁶ Co	⁶⁰ Co
12002,0	B	42 ± 3	75 ± 6	3.5 ± 1.0	13 ± 3	31 ± 12	38 ± 3	33 ± 4	0.55 ± 0.30
12002,20	B	47 ± 3	67 ± 5						0.73 ± 0.65
12002,30	B	86 ± 3	126 ± 6				50 ± 5	148 ± 20	
12004,1	A	53 ± 5	90 ± 6	3.7 ± 1.5			35 ± 4	34 ± 8	< 2.6
12039,0	B	43 ± 5	95 ± 7	< 6.0			37 ± 6	40 ± 10	
12052,1	A	40 ± 6	75 ± 6				27 ± 7	26 ± 10	
12053,0	A	40 ± 6	81 ± 12	7.0 ± 2.0	20 ± 5		35 ± 5	32 ± 6	< 1.0
12054,0	B	39 ± 7	50 ± 10	5.0 ± 2.0			36 ± 5	40 ± 10	< 1.0
12062,0	AB	30 ± 5	57 ± 9	5.0 ± 2.0	9 ± 3		31 ± 6	7 ± 4	
12064,0	B	40 ± 5	51 ± 5	5.0 ± 2.0	22 ± 6	33 ± 18	35 ± 3	32 ± 6	< 1.0
12013,0	C	50 ± 10	115 ± 16	< 15			< 66	50 ± 30	< 8.0
12013,11	C	26 ± 10	90 ± 10						
12034,0	C	29 ± 5	45 ± 5	< 10	< 60		16 ± 8	< 16	< 4.0
12073,0	C	63 ± 7	110 ± 10	< 10			28 ± 7	47 ± 12	
12032,16	D	48 ± 6	100 ± 7	< 10			27 ± 7	< 30	< 2.0
12070,0	D	70 ± 8	146 ± 16				41 ± 10	55 ± 14	< 1.5

* Upper limits are 2 σ evaluated from least-squares analysis.

† A zero following the 5-digit sample number designates a whole rock or fines sample.

‡ Petrologic type according to LSPET (1970).

to determine all 8 nuclides listed in Table 1 for all of the samples. Rock 12002 was the most carefully studied of all our samples and a rather complete radionuclide pattern was obtained. Except for some exceptions noted below, agreement within experimental error was obtained in the few cases where other radionuclide measurements on the same samples could be compared (RANCITELLI *et al.*, 1971; FINKEL *et al.*, 1971).

As was noted previously, ²²Na and ²⁶Al are produced both by solar and galactic cosmic rays (SHEDLOVSKY *et al.*, 1970; PERKINS *et al.*, 1970; O'KELLEY *et al.*, 1970a,b). Because the chemical composition of lunar material favors production of these nuclides, because they can be measured nondestructively by gamma-gamma coincidence methods with high sensitivity, and because their different half-lives (2.6 and 7.4×10^5 years) probe different regions of geologic time, their yields are of great interest.

The concentrations of ²²Na and ²⁶Al from Table 1 may be compared with calculated concentrations produced by galactic protons alone. This permits an estimate of the solar proton component. The ²⁶Al production in a 2π geometry was estimated by the method of FUSE and ANDERS (1969) and the ²²Na production was estimated by the method of BEGEMANN *et al.* (1970). Chemical compositions were taken from the best values from the Apollo 12 Lunar Science Conference and from the Apollo 12 Lunar Sample Catalog (WARNER, 1970).

The comparison between measured and calculated values is shown in Table 2. As a test of the calculations we show in Table 2 data on a sample 10017, ARA which was taken from the bottom of a well-oriented rock, as discussed by O'KELLEY *et al.* (1970b). This bottom piece from 10017 was shielded by about 14 g/cm² of rock, which effectively absorbed the solar protons. Agreement between calculation and experiment is good. Rock 12034 is a breccia recovered from a trench on the north rim of Head Crater; its burial depth was estimated as 15 cm (SHOEMAKER *et al.*, 1970). The low values for the concentrations of ²²Na and ²⁶Al obtained experimentally

Table 2. Comparison between measured concentrations of ^{26}Al and ^{22}Na in lunar rocks and fines compared with concentrations calculated for galactic production only

Sample	²⁶ Al (dpm/kg)		²² Na (dpm/kg)		Remarks
	Measured	Calculated*	Measured	Calculated*	
Crystalline rocks					
10017,ARA	50 ± 7	41	30 ± 5	33	Bottom piece
12002,0	75 ± 6	42	42 ± 3	41	top slice
12002,30	126 ± 6	42	86 ± 3	41	
12004,1	90 ± 6	43	53 ± 5	40	
12052,1	75 ± 6	47	40 ± 6	37	
12053,0	81 ± 12	46	40 ± 6	37	
12062,0	57 ± 9	45	30 ± 5	36	
12064,0	51 ± 5	49	40 ± 5	35	
Breccias					
12013,0	115 ± 16	58	50 ± 10	41	buried 15 cm
12034,0	45 ± 5	52	29 ± 5	40	
12073,0	110 ± 10	50	63 ± 7	38	
Fines					
12032,16	100 ± 7	51	48 ± 6	41	
12070,0	146 ± 16	50	70 ± 8	40	

* Production in 2π geometry by method of FUSE and ANDERS (1969).† Production rates estimated by method of BEGEMANN *et al.* (1970).

show that the rock was shielded from recent solar-proton bombardment. Consideration of the solar-proton spectrum (EBEOGLU and WANIO, 1966; LAL *et al.*, 1967) and the available information on variations in ^{22}Na and ^{26}Al concentrations with depth in lunar materials (FINKEL *et al.*, 1971; RANCITELLI *et al.*, 1971; ELDRIDGE *et al.*, 1971) conservatively specify a burial depth of ≥ 8 cm. Agreement between measured and calculated nuclide concentrations shown in Table 2 for 12034 is also good. It appears that the calculation of BEGEMANN *et al.* (1970) overestimates the ^{22}Na yields slightly.

Samples 12002,30 was a 46-g piece cut from the top of oriented rock 12002,0 and was investigated to obtain depth variations of cosmogenic nuclides by FINKEL *et al.* (1971). Before 12002,30 was submitted to destructive analysis, the data in Table 2 were obtained. As expected, high concentrations of ^{26}Al and ^{22}Na were seen, in excess of the production by galactic protons. It will be noted from Table 1 that the concentration of ^{54}Mn has also been enhanced over the nominal value by the solar proton bombardment while ^{56}Co which is almost totally produced by solar flares manifests a large surface concentration gradient. In contrast, breccia 12034 was shielded from solar protons and shows a low concentration of ^{54}Mn and undetectable ^{56}Co .

For the other rocks of Table 2 large excesses of ^{26}Al over that produced by galactic protons is observed, with moderate excesses of ^{22}Na . These results, together with the ^{56}Co concentrations of Table 1 show that the rocks in question were at least partially exposed on the lunar surface.

Rocks 12054, 12062, and 12064 show evidence of recent low exposure. Within experimental errors it is not possible to decide whether the low values of ^{26}Al are due to partial shielding from solar protons or whether the ^{26}Al did not attain saturation. It will be shown below that galactic proton exposure ages suggest that 12062

and 12064 have been near but not on the lunar surface for the last 150–200 m.y., which may indicate that these rocks received a low exposure to solar protons. Another possible explanation would be a high surface erosion rate, but it is difficult to understand why certain rocks erode rapidly while others do not.

It may be noted that for samples whose radionuclide concentrations can be compared (12062, 12034, 12070) our ^{22}Na concentrations agree with those of RANCITELLI *et al.* (1971) within experimental errors, but our ^{26}Al concentrations appear to be consistently lower.

The two soil samples we examined appear to have been taken from quite different depths. The high concentrations of ^{22}Na and ^{26}Al , and especially the high ^{56}Co , are consistent with near surface sampling for 12070. The sample of 12032 apparently came from a deeper zone, about 5 cm below the surface.

The concentrations of ^{46}Sc in samples from Apollo 12 are about 2.4 times lower than those we found in Apollo 11 samples. This reduction reflects the lower concentration of Ti target nuclei in the samples from the Ocean of Storms.

Despite delays in obtaining samples during the preliminary examination, 5.7-day ^{52}Mn was determined in two rocks and 16-day ^{48}V was determined in four rocks. The ^{52}Mn yields are approximately as expected from the chemical composition and correlate well with the ^{54}Mn yields. Most of the ^{48}V is produced by solar protons via the reaction $^{48}\text{Ti}(p, n)^{48}\text{V}$. To correlate the observed ^{48}V yields with chemical composition, it was necessary to estimate the production of ^{48}V from spallation of iron by high-energy protons. This estimate was derived from the ^{48}V and ^{56}Co concentrations of rock 12062, which showed low exposure to solar-flare protons. By assuming that all ^{56}Co in 12062 was produced by solar-flare protons, the corresponding concentration of ^{48}V was estimated by use of the chemical composition in WARNER (1970) and the (p, n) cross sections for producing ^{48}V and ^{56}Co as measured by TANAKA and FURUKAWA (1959). Of the 9 dpm/kg of ^{48}V shown in Table 1 for 12062, about 3 dpm/kg could be attributed to solar flare production. The 6 dpm/kg of ^{48}V produced by high-energy spallation is not expected to vary significantly among the crystalline rocks of Table 1 because of the nearly constant concentration of iron.

In Fig. 1 we show that the yields of ^{48}V corrected to November 20, 1970, correlate well with the average titanium concentrations reported in the literature for 12002, 12053, and 12064. The flare responsible for the solar ^{48}V occurred on November 3, 1970; if the solar contribution is corrected to that date, the difference between the solid and dashed lines of Fig. 1 will be doubled.

Thermal neutron flux

Cobalt-60 has a half-life of only 5.3 years and is produced with a high cross section (37 barns) by thermal-neutron capture in ^{59}Co . Production of ^{60}Co either by spallation or by the (n, p) reaction in Ni is very low because of the small abundance of the target isotopes and the low cross sections for the nuclear reactions concerned. The concentration of ^{60}Co in lunar material can be employed to calculate the neutron flux characteristic of recent, steady-state production on the lunar surface. Such

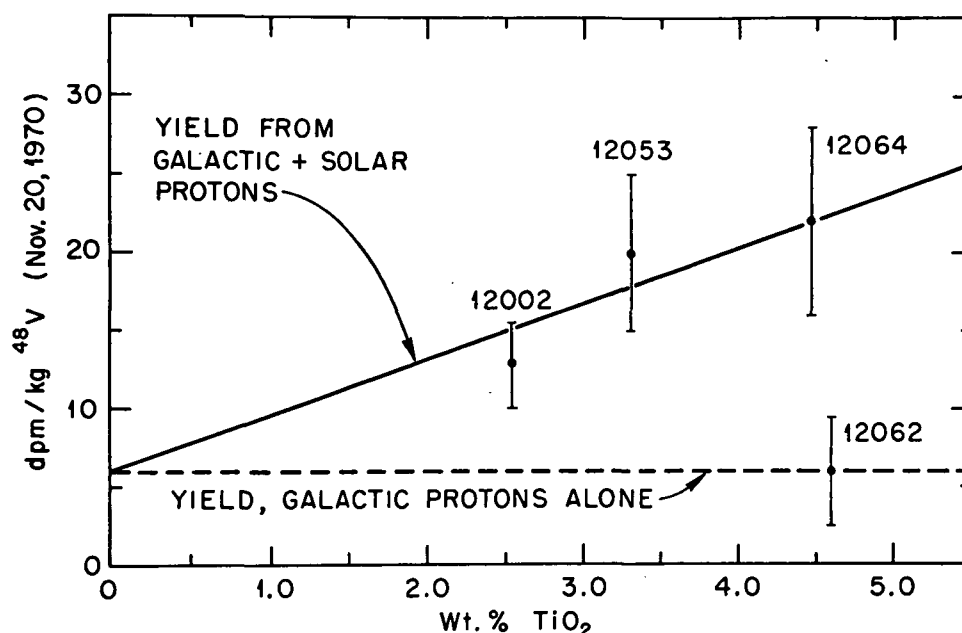


Fig. 1. Correlation between ^{48}V induced by solar-flare protons and titanium concentration. The solid line includes both the yield from solar protons calculated, as described in the text, and the yield from galactic protons.

information is a useful complement to fluxes deduced from mass spectrometric measurements of isotopic anomalies in Gd. The Gd isotope ratios yield an integrated thermal-neutron flux which requires a meaningful exposure age before an average flux can be obtained. Lunar rocks endure such a complex history that the average flux obtained mass spectrometrically may not represent the most recent flux to the accuracy desired.

Although ^{60}Co can be determined in lunar samples by gamma-ray spectrometry, rather large samples are required because the stable ^{59}Co target nuclide is present in such low concentration. Further, the intense interferences from abundant U and Th and cosmogenic radionuclides make difficult the resolution of small quantities of ^{60}Co .

In Table 1 we show that in the case of rock 12002, a value of 0.55 ± 0.30 dpm/kg was obtained for the ^{60}Co concentration. Based on an average Co concentration of 70 ppm in 12002, the thermal-neutron flux was found to be 0.35 ± 0.18 neutrons $\text{cm}^{-2} \text{sec}^{-1}$. The average mass density of rock 12002 was approximately 20 g/cm^2 . Our result for a flux in a 20 g/cm^2 sample is in good agreement with the depth dependence of thermal neutron fluxes measured mass spectrometrically by MARTI and LUGMAIR (1971) in lunar material of about 18 to 150 g/cm^2 . Our result for 12002 is also in agreement with the theoretical value of 0.23 ± 0.06 neutrons $\text{cm}^{-2} \text{sec}^{-1}$ calculated by ARMSTRONG and ALSMILLER (1971), who averaged the solar maximum and mini-

lum fluxes and included nominal Apollo 11 rare-earth concentrations in the lunar surface composition.

Exposure ages

Estimates of cosmic-ray exposure ages were made by the ^{22}Na – ^{22}Ne method as discussed by O'KELLEY *et al.* (1970a, 1970b). It was assumed that the effective cross sections for production of ^{22}Na and ^{22}Ne were equal. Concentrations of Ne were obtained from the literature. The spallogenic ^{22}Ne was estimated to be 1.10 ^{21}Ne . Radioactive concentrations of ^{22}Na were taken from Table 1 and corrected for excess ^{22}Na of solar origin by a semiempirical factor.

In Table 3 we compare our exposure ages from the ^{22}Na – ^{22}Ne method with ^3He exposure ages. The ^3He exposure ages were calculated from a production rate of 10^{-8} cm^3 STP $^3\text{He}/\text{g}$ per 10^6 years exposure. The agreement in Table 3 is gratifying and suggests that the ratio of production rates assumed for the ^{22}Na – ^{22}Ne method is substantially correct.

The rocks of relatively shorter exposure age (12002, 12004, 12013, 12053) all were collected (SUTTON and SCHABER, 1971) in the Ocean of Storms north of a line connecting the north rim of Bench Crater and the center of Surveyor Crater. Rocks 12062, 12064, and 12065 have significantly longer exposure ages and were collected south of this line, which appears to be a boundary associated with Middle Crescent Crater. As shown by WARNER and ANDERSON (1971), most of the crystalline rocks north of this diffuse boundary are porphyritic basalts, while those to the south are granular and ophitic basalts. The model proposed by WARNER and ANDERSON to account for this distribution tentatively associated with Middle Crescent Crater suggests that the area north of the boundary would be strewn with ejecta of somewhat more recent exposure than the region to the south, which might be rich in older regolith material. Although this conclusion is speculative and is based on relatively few exposure ages, our data lend qualitative support to the WARNER and ANDERSON model.

Table 3. Estimation of exposure ages of Apollo 12 lunar samples.

Sample	Exposure age (10^6 y)		Ref. gas data
	^3He	^{22}Na – ^{22}Ne	
12002	89	96	a
12004	61	58	b
12013	40	48	c
12053	79	99	b
12062	150	153	b
12064	205	251	b
12065	182	217	b

a. ^3He from HINTENBERGER *et al.* (1971); ^{21}Ne from MARTI and LUGMAIR (1971).

b. ^3He and ^{21}Ne concentrations from HINTENBERGER *et al.* (1971).

c. ^3He and ^{21}Ne concentrations from SCHAEFFER *et al.* (1970).

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REFERENCES

- ARMSTRONG T. W. and ALSMILLER R. G., JR. (1971) Calculation of cosmogenic radionuclides in the moon and comparison with Apollo measurements. Second Lunar Science Conference (unpublished proceedings).
- BEGEMANN, F., VILCSEK E., RIEDER R., BORN W., and WÄNKE H. (1970) Cosmic-ray produced radioisotopes in lunar samples from the Sea of Tranquillity (Apollo 11). *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 995–1007. Pergamon.
- EBEOGLU D. B. and WAINIO K. M. (1966) Solar proton activation of the lunar surface. *J. Geophys. Res.* 71, 5863–5872.
- ELDRIDGE J. S., O'KELLY G. D., SCHONFELD E., and NORTH CUTT K. J. (1971) Unpublished data.
- FINKEL R. C., ARNOLD J. R., REEDY R. C., FRUCTER J. S., LOOSLI H. H., EVANS J. C., SHEDLOVSKY J. P., IMAMURA M., and DELANY A. C. (1971) Depth variation of cosmogenic nuclides in a lunar surface rock. Second Lunar Science Conference (unpublished proceedings).
- FUSE K. and ANDERS E. (1969) Aluminum-26 in meteorites. VI. Achondrites. *Geochim. Cosmochim. Acta* 33, 653–670.
- HERZOG G. F. and HERMAN G. F. (1970) Na²², Al²⁶, Th and U in Apollo 11 lunar samples. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1239–1247. Pergamon.
- HINTENBERGER H., WEBER H., and TAKAOKA N. (1971) Concentrations and isotopic abundances of the rare gases in lunar matter. Second Lunar Science Conference (unpublished proceedings).
- LAL D., RAJAN R. S., and VENKATAVARADAN V. S. (1967) Nuclear effects of "solar" and "galactic" cosmic-ray particles in near-surface regions of meteorites. *Geochim. Cosmochim. Acta* 31, 1859–1869.
- LSPET (Lunar Sample Preliminary Examination Team) (1970) Preliminary examination of lunar samples from Apollo 12. *Science* 167, 1325–1339.
- MARTI K. and LUGMAIR G. W. (1971) Kr⁸¹–Kr and K–Ar⁴⁰ ages, cosmic-ray spallation products and neutron effects in Apollo 11 and Apollo 12 lunar samples. Second Lunar Science Conference (unpublished proceedings).
- O'KELLY G. D., ELDRIDGE J. S., SCHONFELD E., and BELL P. R. (1970a) Elemental compositions and ages of lunar samples by nondestructive gamma-ray spectrometry. *Science* 167, 580–582.
- O'KELLY G. D., ELDRIDGE J. S., SCHONFELD E., and BELL P. R. (1970b) Primordial radionuclide abundances, solar-proton and cosmic-ray effects and ages of Apollo 11 lunar samples by non-destructive gamma-ray spectrometry. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1407–1423. Pergamon.
- O'KELLY G. D., ELDRIDGE J. S., SCHONFELD E., and BELL P. R. (1971) Abundances of the primordial radionuclides K, Th and U in Apollo 12 lunar samples by nondestructive gamma-ray spectrometry: Implications for origins of lunar soils. Second Lunar Science Conference (unpublished proceedings).
- PERKINS R. W., RANCITELLI L. A., COOPER J. A., KAYE J. H., and WOGMAN N. A. (1970) Cosmogenic and primordial radionuclide measurements in Apollo 11 lunar samples by nondestructive analysis. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1455–1471. Pergamon.
- RANCITELLI L. A., PERKINS R. W., FELIX W. D., and WOGMAN N. A. (1971) Cosmogenic and primordial radionuclide measurements in Apollo 12 lunar samples by nondestructive analysis. Second Lunar Science Conference (unpublished proceedings).

- SCHONFELD E. and O'KELLEY G. D. (1971) The selenographic orientation of Apollo 12 rocks determined by nondestructive gamma-ray spectroscopy. Second Lunar Science Conference (unpublished proceedings).
- SCHAEFFER O. A., FUNKHOUSER J. G., BOGARD D. D., and ZÄRINGER J. (1970) Potassium-argon ages of lunar rocks from Mare Tranquillitatus and Oceanus Procellarum. *Science* **170**, 161-162.
- SHEDLOVSKY J. P., HONDA M., REEDY R. C., EVANS J. C., JR., LAL D., LINDSTROM R. M., DELANY A. C., ARNOLD J. R., LOOSLI H. H., FRUCHTER J. S., and FINKEL R. C. (1970) Pattern of bombardment-produced radionuclides in rock 10017 and in lunar soil. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1503-1533. Pergamon.
- SHOEMAKER E. M., BATSON R. M., BEAN A. L., CONRAD C. JR., DAHLEM D. H., GODDARD E. N., HAIT M. H., LARSON K. B., SCHABER G. G., SCHLEICHER D. L., SUTTON R. L., SWANN G. A., and WATERS A. C. (1970) Geology of the Apollo 12 landing site. In *Apollo 12 Preliminary Science Report, NASA SP-236*, NASA Manned Spacecraft Center, Houston, pp. 113-156.
- SUTTON R. L. and SCHABER G. G. (1971) Lunar locations and orientations of rock samples from Apollo missions 11 and 12. Second Lunar Science Conference (unpublished proceedings).
- TANAKA S. and FURUKAWA M. (1959) Excitation functions for (*p*, *n*) reactions with titanium, vanadium, chromium, iron and nickel up to $E_p = 14$ MeV. *J. Phys. Soc. Japan*, **14**, 1269-1275.
- WARNER J. L., Compiler (1970) *Apollo 12 lunar sample information*. NASA Manned Spacecraft Center report S-243, Houston, Texas.
- WARNER J. L. and ANDERSON D. H. (1971) Lunar crystalline rocks: Petrology, geology and origin. Second Lunar Science Conference (unpublished proceedings).
- WRIGLEY R. C. and QUAIDE W. L. (1970) Al^{26} and Na^{22} in lunar surface materials: Implications for depth distribution studies. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1751-1757. Pergamon.

Abundances of the primordial radionuclides K, Th, and U in Apollo 12 lunar samples by nondestructive gamma-ray spectrometry: Implications for origin of lunar soils

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Abstract—Gamma-ray spectrometers with low background were used to determine concentrations of K, Th, and U in crystalline rocks 12002, 12004, 12021, 12039, 12051, 12052, 12053, 12054, 12062, 12064, and 12065; in breccias 12013, 12034, and 12073; and in fines samples 12032 and 12070. The crystalline rocks studied resemble the low-potassium rocks of Apollo 11 and have average concentrations of K, Th, and U of 520, 0.97, and 0.25 ppm, respectively, with remarkably small deviations from the average values. Concentration ratios K/U for all Apollo 11 and Apollo 12 materials vary only from 1200 to 3200 for a range of 50 in K or U concentration. With the body of data now available on samples from Apollo 11 and Apollo 12, it appears that there are five groups of materials with characteristic K/U ratios.

The Apollo 12 fines and breccia together are relatively uniform, but, because of their much higher concentrations of radioactive elements, they could not have been formed directly from the crystalline rocks examined. Rock 12013 has the highest concentrations of K, Th, and U, but the lowest ratio Th/U of any lunar sample we have studied. Our systematics of primordial radioelement concentrations predict some of the properties of the foreign component of the lunar soil and breccia. It is suggested that the dark portion of 12013 contains a high concentration of this foreign material.

Concentrations of K, Th, and U obtained in this study were combined with gas analysis data from other workers to obtain K-⁴⁰Ar ages from 2.34 to 3.37 aeons and U, Th-⁴He ages from 1.22 to 2.69 aeons.

INTRODUCTION

DATA ON THE abundances of the primordial radioelements K, Th, and U are essential to many studies of geological processes. Such information is of great importance in formulating theories of planetary evolution where, because of their radioactivity, the contribution of primordial radionuclides to the thermal balance of the planet must be considered. The concentration of K can serve as a useful indicator for alkali-metal chemistry. Further, since K is a relatively volatile element and Th and U are refractory, concentrations of these elements may be invoked to establish limits on a temperature profile for a planetary body or other geological system under study.

Gamma-ray spectrometry has proved to be a very useful method for determination of K via its radioactive isotope ⁴⁰K, and U and Th via their respective decay products. Studies on lunar samples from Apollo 11 demonstrated that nondestructive, gamma-ray spectrometry measurements could be made with good accuracy on samples which

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varied in size from a few grams to kilograms (O'KELLY *et al.*, 1970a, 1970b; PERKINS *et al.*, 1970; HERZOG and HERMAN, 1970; WRIGLEY and QUAIDE, 1970).

Because of the widespread geochemical interest in concentrations of K, Th, and U, and because the experimental method is rapid and nondestructive, it was deemed useful to scan a large number of lunar samples by gamma-ray spectrometry. The measurements reported here show interesting geochemical trends for the Apollo 12 surface materials which are compared to a less extensive survey of Apollo 11 materials. In addition, this survey yielded the first evidence that rock 12013 was dramatically different in its chemistry from all lunar samples previously examined.

Preliminary data on some of the samples examined in this study during the Apollo 12 mission were reported in LSPET (1970). Since the publication of the preliminary study, the data analyses have been refined and additional samples have been analyzed.

EXPERIMENTAL METHODS

The gamma scintillation coincidence spectrometer which was used at the Lunar Receiving Laboratory (LRL) for analysis of most of the samples reported in this study contained two NaI(Tl) detectors, each 23 cm in diameter and 13 cm long, with 10 cm pure NaI light guides. This spectrometer system and associated data reduction techniques were described previously by O'KELLY *et al.* (1970b). Stainless steel cans of 0.8 mm wall thickness, 16 cm in diameter and either 5.6 or 7.6 cm in height were used as sample containers during the quarantine period; later, thin teflon bags were used to protect the samples during measurements after quarantine restrictions were removed.

A second detector system, identical to that at the LRL and located at Oak Ridge National Laboratory (ORNL), was used to analyze two of the samples (12002,20 and 12013,11) reported in this study. The ORNL data acquisition system used a 4096-channel coincidence analyzer and was operated in a 64×64 channel matrix configuration with both energy axes adjusted to 60 keV/channel to cover an energy range of 0–3.8 MeV in each detector. An IBM 360/91 program was used to pre-process the matrix data and separate the spectra into singles spectra from the two detectors, a summed singles spectrum from the two detectors, and gamma-gamma coincidence spectra from selected regions of the matrix folded in a manner similar to that which we described for the LRL system (O'KELLY *et al.*, 1970b). The series of single-parameter spectra obtained in this way were plotted and then were analyzed by use of ALPHA-M, a computer program for quantitative radio-nuclide determination by the method of least squares (SCHONFELD, 1967). Analyses carried out with the ORNL system were in excellent agreement with those made with the LRL system.

For analyses of data recorded during the preliminary study and reported in LSPET (1970), calibration of the LRL coincidence spectrometer was established by recording a library of spectra from cylindrical radioactive standards prepared by dispersing known amounts of radioactivity in quantities of iron powder. Absorption factors to correct the data for the presence of the stainless steel containers were computed. These corrections were especially important for the cosmogenic radionuclide data, relatively small for the U and Th determinations, and least significant for the ^{40}K data. The data reported here were obtained by using, when required, a new library of spectra recorded with the standard sources placed inside the stainless steel containers actually used.

Spectrum libraries employed in the analysis of samples 12002,0; 12002,20; 12013,11; 12032,16; 12034,0; 12070,0; and 12073,0 were obtained from replicas which accurately reproduced the electronic and bulk densities of the lunar samples. Procedures for preparation of the cylindrical standards and the replicas were described earlier by O'KELLY *et al.* (1970a, 1970b).

Error statements given in Table 1 are quite conservative estimates of the overall uncertainties, including counting statistics, geometrical factors, and calibration errors. A detailed analysis of a low-level synthetic lunar sample containing eight components showed that our methods were capable of absolute uncertainties $\leq 2.5\%$ for samples of the same degree of complexity as the samples studied here (O'KELLY *et al.*, 1970b).

Table 1. Concentrations of primordial radionuclides in lunar samples*

Sample	Weight (g)	K (ppm)	Th (ppm)	U (ppm)	K/U Mass ratio	Th/U Mass ratio
<i>Crystalline rocks</i>						
12002,0	1529	450 ± 20	0.89 ± 0.06	0.23 ± 0.02	1960 ± 192	3.87 ± 0.42
12002,20	260	425 ± 21	0.73 ± 0.04	0.21 ± 0.02	2020 ± 216	3.50 ± 0.39
12002,30	46	440 ± 20	0.86 ± 0.06	0.22 ± 0.02	2000 ± 212	3.91 ± 0.45
12004,1	502	469 ± 33	0.92 ± 0.09	0.24 ± 0.03	1960 ± 282	3.83 ± 0.61
12021,0	1877	500 ± 50	0.98 ± 0.10	0.26 ± 0.03	1920 ± 292	3.77 ± 0.53
12039,0	255	673 ± 40	1.20 ± 0.06	0.31 ± 0.03	2170 ± 246	3.87 ± 0.42
12051,0	1660	530 ± 50	1.00 ± 0.10	0.26 ± 0.03	2040 ± 304	3.85 ± 0.59
12052,1	201	540 ± 20	1.03 ± 0.06	0.27 ± 0.02	2000 ± 166	3.82 ± 0.36
12053,0	879	535 ± 40	1.06 ± 0.11	0.28 ± 0.03	1910 ± 244	3.79 ± 0.57
12054,0	687	530 ± 35	0.79 ± 0.08	0.22 ± 0.03	2410 ± 365	3.59 ± 0.61
12062,0	739	510 ± 35	0.83 ± 0.09	0.22 ± 0.03	2320 ± 354	3.78 ± 0.66
12064,0	1214	520 ± 35	0.87 ± 0.09	0.23 ± 0.02	2260 ± 248	3.78 ± 0.51
12065,0	2109	510 ± 50	1.06 ± 0.11	0.27 ± 0.03	1890 ± 280	3.92 ± 0.54
Average, crystalline rocks					2066	3.79 ± 0.14
<i>Breccia</i>						
12034,0	155	4560 ± 130	13.1 ± 0.3	3.4 ± 0.2	1341 ± 88	3.85 ± 0.24
12073,0	405	2960 ± 90	8.45 ± 0.10	2.19 ± 0.08	1352 ± 68	3.86 ± 0.15
<i>Fines</i>						
12032,16	89.6	3100 ± 100	8.8 ± 0.2	2.35 ± 0.07	1320 ± 58	3.75 ± 0.14
12070,0	354	2030 ± 120	6.25 ± 0.50	1.65 ± 0.16	1230 ± 140	3.79 ± 0.48
Average, fines and breccia					1311	3.81 ± 0.14

* Standardization for assay of K, Th, and U assumed terrestrial isotopic abundances and equilibrium of Th and U decay series.

RESULTS AND DISCUSSION

Analysis of rocks and fines

Concentrations of K, Th, and U in 11 crystalline rocks, two breccias, and two samples of fines are listed in Table 1. The large number of samples studied permits some detailed comparisons to be made between the concentrations of Table 1 and comparable measurements carried out on Apollo 11 samples by several gamma-ray spectrometry teams (O'KELLEY *et al.* 1970a, 1970b; PERKINS *et al.* 1970; HERZOG and HERMAN, 1970; WRIGLEY and QUAIDE, 1970).

The crystalline rocks of Apollo 11 appeared to fall into two chemical groups with different alkali metal concentrations. Our Apollo 11 work showed that one chemical group had average K, Th, and U concentrations of about 500, 1.0, and 0.25 ppm, respectively; the other group had respective average K, Th, and U concentrations of about 2400, 3.3, and 0.8 ppm. Similar chemical differences were noted by GAST *et al.* (1970) and by COMPSTON *et al.* (1970).

All of the crystalline rocks of Apollo 12 which we have examined resemble the low-potassium group of Apollo 11 and have average concentrations of K, Th, and U of about 520, 0.97, and 0.25 ppm, respectively. These Apollo 12 samples show remarkably small deviations from the average values. WARNER and ANDERSON (1971) have suggested that the crystalline rocks of Oceanus Procellarum can be classified into petrographic groups and some attempts have been made to classify these Apollo 12 basalts into various chemical groups. However, the chemical differences are quite

subtle (e.g., GOLES *et al.*, 1971) and more data are needed to compare the petrographic and chemical classifications.

The fines and breccia from the Ocean of Storms are quite different from the crystalline rocks in several respects. The concentrations of the radioactive elements K, Th, and U are much higher and more variable in the fines and breccia than in the crystalline rocks. The average ratio K/U for fines and breccia is only about 1300 (see Table 1), compared with an average value of about 2100 for the crystalline rocks. It is important to note that the mass ratio Th/U is approximately 3.8 for all materials listed in Table 1, in good agreement with average Th/U ratios for Tranquillity Base materials and terrestrial materials. The concentrations of K, U, and Th in the fines and breccia from the Apollo 11 site were intermediate between the respective concentrations for the two chemical classes of crystalline rocks. It appeared on this basis that the fines and breccia might have been derived from the crystalline rocks at the Sea of Tranquillity; however, more detailed chemical studies later showed that Apollo 11 soil contained other components differing slightly from the crystalline rocks in composition (WOOD *et al.*, 1970; WASSON *et al.*, 1970). Clearly, the crystalline rocks whose concentrations are given in Table 1 could not alone have formed the Apollo 12 fines and breccia.

Preliminary observations that the crystalline lunar rocks are strongly depleted in volatile elements (LSPET 1969, 1970) have been substantiated by detailed studies (e.g., GANAPATHY *et al.*, 1970a, 1970b). This depletion of volatile elements is thought to be characteristic of the whole moon. Studies of the primordial radioactive elements are particularly useful in this connection, because the ratios of K (a volatile element) to U (a refractory element) for common terrestrial rock types are remarkably constant over a wide range of K concentration. Thus, the K/U ratio appears to be a result of early chemical fractionation and is not affected by later igneous processes. It was noted by WASSERBURG *et al.*, (1964) that the terrestrial average K/U ratio of 10^4 was distinct from K/U ratios for meteorites.

In Fig. 1 we show a plot of K/U mass ratios as a function of potassium concentration, normalized to 18 wt. % silicon. Data on lunar samples are from our gamma spectrometry studies and from other chemical analyses reported in the literature. It is clear that eucrites, carbonaceous chondrites, and ordinary chondrites fall into groups which are distinct from one another and from either lunar or terrestrial materials. It may be noted that on these systematics the tektites fall within the terrestrial group. The meteorites fall on a common "trend line" for which the product of the K and U concentrations is constant. In contrast, both the earth and the moon possess constant K/U ratios characteristic of each planet.

The values of K/U for lunar material appear to be separable into five distinct groups, as shown in Fig. 1: (a) Apollo 12 rocks + low-K Apollo 11 rocks; (b) Apollo 11 fines and breccia; (c) high-K Apollo 11 rocks; (d) Apollo 12 fines and breccia; and (e) rock 12013. The relationships between these "islands" in the K/U systematics suggest some interesting possibilities. If trend lines were drawn in Fig. 1 through the Apollo 11 points and through the Apollo 12 data (excluding rock 12013), the intersection falls in the group of points for the crystalline rocks of Apollo 12 and the low-K rocks of Apollo 11. It may be significant that this intersection falls near the extra-

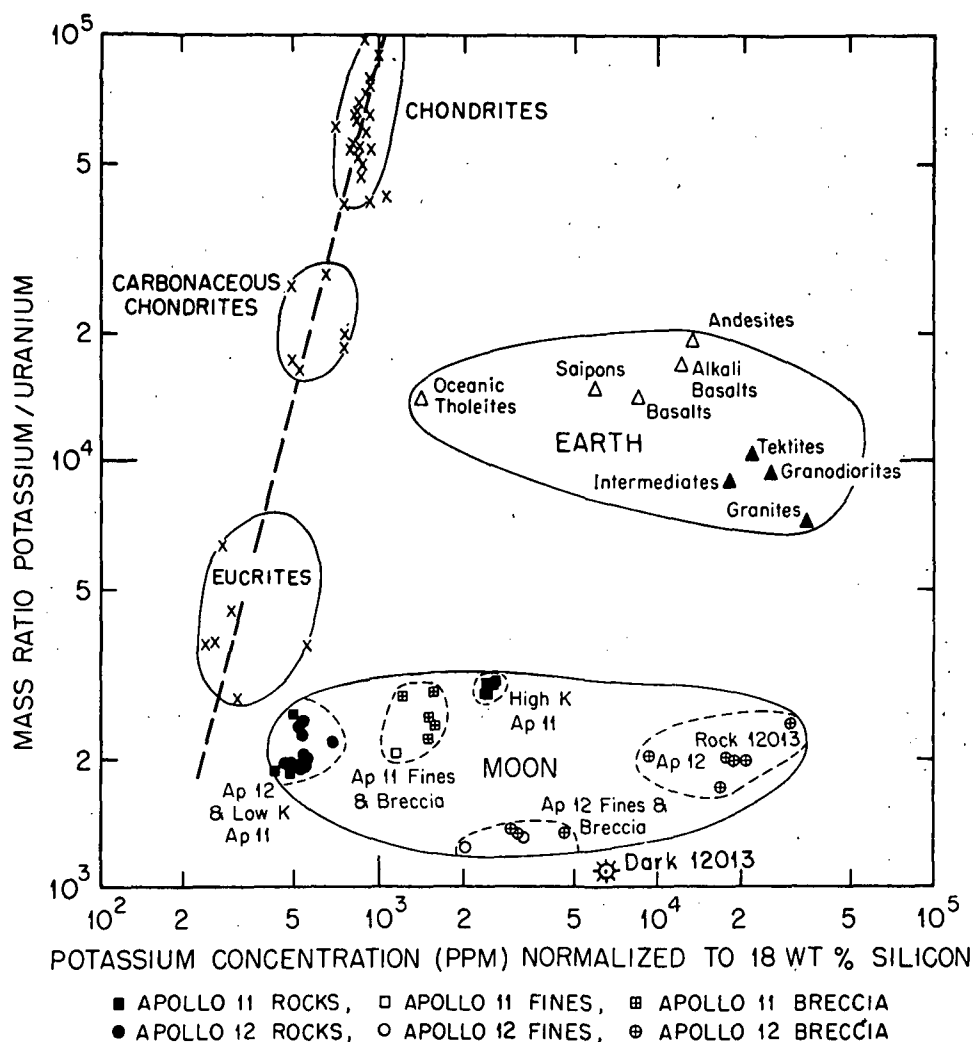


Fig. 1. Plot of mass ratios K/U as a function of K concentration normalized to 18 wt. % Si. The distinct grouping of meteorites and terrestrial and lunar materials is apparent.

polated trend line for meteorites. The divergence of the trend lines for Apollo 11 and Apollo 12 materials from each other suggests that the samples at the two landing sites are the products of two separate magmatic processes which originated from material with similar K and U concentrations. This conclusion was reached by FANALE and NASH (1971) who analyzed preliminary K and U data by a different graphical method. This analysis also implies that the white portion of rock 12013 was involved in a third magmatic event. The starting material for all three events is depleted in K but enriched in U with respect to chondrites.

Rock 12013 and the origin of Apollo 12 fines and breccia

Sample 12013 proved to be one of the most interesting samples studied. Our gamma-ray spectrometry measurements during the preliminary examination showed clearly that the specimen was dramatically different from all lunar samples previously examined (LSPET, 1970). The concentrations of K and U were about 40 times the average concentrations of the crystalline rocks of Apollo 12 and about 10 times the concentrations of the high-K rocks of Apollo 11. In Table 2 we show a refined analysis of our data on 12013,0 and an additional measurement on the 66-g piece (12013,11) which remained after samples were cut for destructive analysis. Despite the inhomogeneity of this breccia on a microscopic scale, the primordial radionuclide concentrations from our two measurements on large rock fragments agree very well with each other. The Th/U ratio is distinctly different, namely, 3.3 compared with 3.8 for other Apollo 12 material.

It is interesting to speculate on the possible role of rock 12013 or one of its components in forming the Apollo 12 fines and breccia. Our K/U systematics in Fig. 1 suggest that the required material to mix with the crystalline rocks must have a low ratio K/U of about 1000 and a K concentration of about 5000–7000 ppm. Further, since our Th/U ratios of Table 1 show that the fines, breccia, and crystalline rocks all have a common Th/U ratio of 3.8, not all of the constituents of 12013 can be common to the lunar soil.

Studies on 12013 have disclosed a general chemical similarity between the light and gray material, which is distinct from the dark material. Chemical analyses on these two classes of components were averaged with the results shown in Table 2. The average K concentration of 6500 ppm for the dark material and 25,500 ppm for the light and gray is in fair agreement with our measured concentration of about 20,000 ppm for the whole rock. The K concentration of the light material is too high to be the missing component. Further, WAKITA and SCHMITT (1970) report a range of 2.3–3.2 for the Th/U ratio, which also militates against the light material. However, TATSUMOTO (1970) found dark material from 12013,10 which had a U concentration of 5.7 ppm and a “normal” Th/U mass ratio of 3.7. Thus, it appears that a component of

Table 2. Concentrations of K, Th, and U in lunar rock 12013.

Sample	Weight (g)	K (ppm)	Th (ppm)	U (ppm)	K/U Mass ratio	Th/U Mass ratio
12013,0*	80.0	20,400 ± 600	34.2 ± 0.8	10.3 ± 0.5	1980 ± 112	3.32 ± 0.18
12013,11*	66.2	21,100 ± 600	32.2 ± 1.4	9.8 ± 1.0	2160 ± 228	3.29 ± 0.36
12013,10 (dark)	—	~6500†		~5.7‡	~1100	3.7‡
12013,10 (light)	—	25,500§		~13.0	~2000	(2.3–3.2)

* Standardization for assay of K, Th, and U assumed terrestrial isotopic abundances and equilibrium of Th and U decay series.

† Average of analyses by SCHNETZLER *et al.* (1970), HUBBARD *et al.* (1970) and TURNER (1970).

‡ Estimated from TATSUMOTO (1970).

§ Average of analyses by ALEXANDER (1970), SCHNETZLER *et al.* (1970), HUBBARD *et al.* (1970), TURNER (1970), and WAKITA and SCHMITT (1970).

|| WAKITA and SCHMITT (1970).

12013 with a K concentration of 6500 ppm, a K/U ratio of ~ 1100 , and a Th/U ratio of 3.8 does exist and may be the component required to form the lunar fines and breccia from the crystalline rocks. This point has been added to the K/U systematics of Fig. 1.

These results strongly support the suggestions of HUBBARD *et al.* (1970) and SCHNETZLER *et al.* (1970) that the dark component of 12013 may be a major component of the lunar soil. However, as recently proposed by HUBBARD *et al.* (1971), the dark material probably contains in turn the exotic basaltic material high in K, rare earth elements and P (KREEP), which appears to comprise a large percentage of the lunar soil. If the KREEP material is found to be high in uranium as discussed above, it would serve the purpose of our K/U systematics equally well.

In Fig. 2 we show two-component mixing diagrams which are in accord with the requirements of our K/U systematics and with the properties of the dark component of 12013. The K mixing line is fitted to our average K concentration of 520 ppm for lunar basalts and to 6500 ppm for the foreign component of the soil. The U mixing line is required to intercept the left ordinate at our average concentration for lunar basalts of 0.25 ppm; however, the intercept on the right ordinate was determined by the best fit of a straight line to all data points. On this basis our best estimate of the U concentration in the foreign component is 5.2 ppm. The agreement obtained for five sets of data is very good for such a simple model. Although concentrations of Al, Mg, and Fe in the crystalline rocks vary over a rather wide range, HUBBARD *et al.* (1971) found that the two-component mixing model for Apollo 12 soils also could explain adequately the variations in the concentrations of the major elements Si, Al, Mg, Ti, and Fe.

Gas retention ages

The elemental analyses for K, Th, and U reported in Table 1 may be combined with rare gas concentrations to estimate crystallization ages of rocks. In Table 3 we show

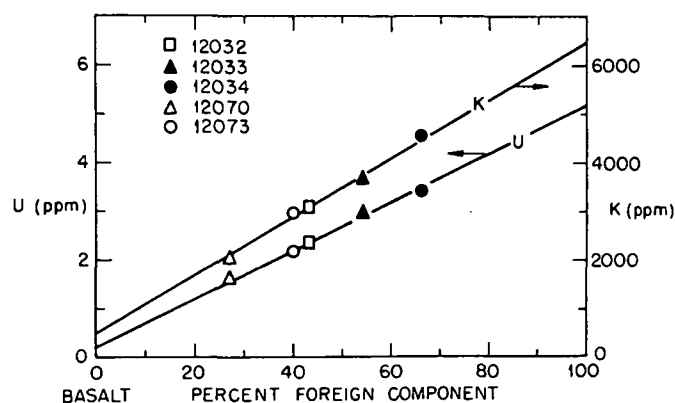


Fig. 2. Two-component mixing lines for K and U concentrations in lunar soils and breccia. Samples (and per cent foreign component) are: soils 12032 (43%), 12033 (54%), 12070 (27%); and breccia 12034 (66%), 12073 (40%).

Table 3. Estimation of gas retention ages of lunar samples.

Sample	K- ⁴⁰ Ar (10 ⁹ y)	U, Th- ⁴ He (10 ⁹ y)
12002	2.55*	2.32*
12004	2.92*	2.69*
12021	3.22†	
12052	2.34‡	
12062	2.61‡	1.22§
12064	3.10*	2.69*
12065	2.79*	1.95*
12013	3.37‡	1.47‡

* Rare gas concentration from HINTENBERGER *et al.* (1971); K, Th, U concentrations from Table 1.

† Rare gas concentrations from MARTI and LUGMAIR (1971); K, Th, U concentrations from Table 1.

‡ Rare gas concentrations from LSPET (1970) and from SCHAEFFER *et al.* (1970); K, Th, U concentrations from Table 1.

§ Rare gas concentrations from LSPET (1970); Th and U concentrations from Table 1.

gas retention ages for 8 samples. Although the K-⁴⁰Ar and U, Th-⁴He ages are approximately concordant for 12002, 12004, 12021, and 12064, the U, Th-⁴He ages are consistently younger than the K-⁴⁰Ar ages. The low U, Th-⁴He ages for 12062, 12065, and 12013 suggest that these rocks have experienced significant heating accompanied by preferential loss of He.

Gas retention ages of large samples are generally found to give somewhat shorter ages than those obtained by other methods. However, the K-⁴⁰Ar ages of Table 3 for 12004, 12021, and 12064 show evidence of little or no loss of radiogenic argon. These ages compare well with the range for all Apollo 12 rocks reported by ALBEE *et al.* (1971) to lie between 3.15 and 3.35 b.y. Our value for 12064 of 3.10 b.y. lies within experimental error of this range. Our value of 3.22 b.y. for 12021 is in good agreement with 3.28 b.y. obtained by CLIFF *et al.* (1971). Similarly, our value of 2.92 b.y. for 12004 agrees with 3.00 b.y. by MURTHY *et al.* (1971). None of the Apollo 12 rocks we have studied have exhibited an age as high as the 3.97 b.y. we determined for Apollo 11 rock 10003 (O'KELLY *et al.*, 1970b).

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REFERENCES

- ALBEE A. L., BURNETT D. S., CHODOS A. A., HAINES E. L., HUNEKE J. C., PODOSEK F. A., PAPANASTASSIOU D. A., PRICE G., TERA F., and WASSERBURG G. J. (1971). Rb-Sr ages, chemical abundance patterns and history of lunar rocks. Second Lunar Science Conference (unpublished proceedings).
- ALEXANDER E. C., JR. (1970). Rare gases from stepwise heating of lunar rock 12013. *Earth Planet. Sci. Lett.* 9, 201-208.
- COMPSTON W., CHAPPELL B. W., ARRIENS P. A., and VERNON M. J. (1970). The chemistry and age of Apollo 11 lunar material. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1007-1029. Pergamon.

- CLIFF R. A., LEE-HU C., and WETHERILL G. W. (1971). Rb-Sr and U, Th-Pb measurements on Apollo 12 material. Second Lunar Science Conference (unpublished proceedings).
- FANALE F. P. and NASH D. B. (1971). Potassium-uranium systematics of Apollo 11 and Apollo 12 samples: Implications for lunar material history. *Science* **171**, 282-284.
- GANAPATHY R., KEAYS R. R., LAUL J. C., and ANDERS E. (1970a). Trace elements in Apollo 11 lunar rocks: Implications for meteorite influx and origin of moon. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 117-1143. Pergamon.
- GANAPATHY R., KEAYS R. R., and ANDERS E. (1970b). Apollo 12 lunar samples: Trace element analysis of a core and the uniformity of the regolith. *Science* **170**, 533-535.
- GAST P. W., HUBBARD N. J., and WIESMANN H. (1970). Chemical composition and petrogenesis of basalts from Tranquillity Base. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1143-1165. Pergamon.
- GOLES G. G., DUNCAN A. R., OSAWA M., MARTIN M. R., BEYER R. L., LINDSTROM D. J., and RANDLE K. (1971). Analyses of Apollo 12 specimens and a mixing model for Apollo 12 soils. Second Lunar Science Conference (unpublished proceedings).
- HERZOG G. F. and HERMAN G. F. (1970). Na²², Al²⁸, Th, and U in Apollo 11 lunar samples. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1239-1247. Pergamon.
- HINTENBERGER H., WEBER H., and TAKAOKA N. (1971). Concentrations and isotopic abundances of the rare gases in lunar matter. Second Lunar Science Conference (unpublished proceedings).
- HUBBARD N. J., GAST P. W., and WIESMANN H. (1970). Rare earth, alkaline and alkali metal and ^{87/86}Sr data for subsamples of lunar sample 12013. *Earth Planet. Sci. Lett.* **9**, 181-184.
- HUBBARD N. J., MEYER C., GAST P. W., and WIESMANN H. (1971). The composition and derivation of Apollo 12 soils. *Earth Planet. Sci. Lett.* **10**, 341-350.
- LSPET (Lunar Sample Preliminary Examination Team) (1970). Preliminary examination of lunar samples from Apollo 11. *Science* **165**, 1211-1227.
- LSPET (Lunar Sample Preliminary Examination Team) (1970). Preliminary examination of lunar samples from Apollo 12. *Science* **167**, 1325-1339.
- MARTI K. and LUGMAIR G. W. (1971). Kr⁸¹-Kr and K-Ar⁴⁰ ages, cosmic-ray spallation products and neutron effects in Apollo 11 and Apollo 12 lunar samples. Second Lunar Science Conference (unpublished proceedings).
- MURTHY V. R., EVENSEN N. M., JAHN B., and COSCIO M. R. (1971). Rb-Sr isotopic relations and elemental abundances of K, Rb, Sr and Ba in Apollo 11 and Apollo 12 samples. Second Lunar Science Conference (unpublished proceedings).
- O'KELLEY G. D., ELDRIDGE J. S., SCHONFELD E., and BELL P. R. (1970a). Elemental compositions and ages of lunar samples by non-destructive gamma-ray spectrometry. *Science* **167**, 580-582.
- O'KELLEY G. D., ELDRIDGE J. S., SCHONFELD E., and BELL P. R. (1970b). Primordial radionuclide abundances, solar-proton and cosmic-ray effects and ages of Apollo 11 lunar samples by non-destructive gamma-ray spectrometry. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1407-1423. Pergamon.
- PERKINS R. W., RANCITELLI L. A., COOPER J. A., KAYE J. H., and WOGMAN N. A. (1970). Cosmogenic and primordial radionuclide measurements in Apollo 11 lunar samples by nondestructive analysis. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1455-1471. Pergamon.
- SCHONFELD E. (1967). ALPHA M— an improved computer program for determining radioisotopes by least-squares resolution of the gamma-ray spectra. *Nucl. Instrum. Methods* **52**, 177-178.
- SCHAEFFER O. A., FUNKHOUSER J. G., BOGARD D. D., and ZÄRINGER J. (1970). Potassium-argon ages of lunar rocks from Mare Tranquillitatis and Oceanus Procellarum. *Science* **170**, 161-162.
- SCHNETZLER C. C., PHILPOTTS J. A., and BOTTINO M. L. (1970). Li, K, Rb, Sr, Ba and rare-earth concentrations, and Rb-Sr age of lunar rock 12013. *Earth Planet. Sci. Lett.* **9**, 185-192.
- TATSUMOTO M. (1970). U-Th-Pb age of Apollo 12 rock 12013. *Earth Planet. Sci. Lett.* **9**, 193-200.
- TURNER G. (1970). ⁴⁰Ar-³⁹Ar age determination of lunar rock 12013. *Earth Planet. Sci. Letters* **9**, 177-181.
- WAKITA H. and SCHMITT R. A. (1970). Elemental abundances in seven fragments from lunar rock 12013. *Earth Planet. Sci. Lett.* **9**, 169-176.

- WARNER J. L. and ANDERSON D. L. (1971). Lunar crystalline rocks: petrology, geology and origin. Second Lunar Science Conference (unpublished proceedings).
- WASSERBURG G. J., MACDONALD G. J. F., HOYLE F., and FOWLER W. A. (1964). Relative contributions of uranium, thorium, and potassium to heat production in the earth. *Science* **143**, 465-467.
- WASSON J. T. and BAEDECKER P. A. (1970). Ga, Ge, In, Ir, and Au in lunar, terrestrial, and meteoritic basalts. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1741-1750. Pergamon.
- WOOD J. A., DICKEY J. S., Jr., MARVIN U. B., and POWELL B. N. (1970). Lunar anorthosites and a geophysical model of the moon. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 1, pp. 965-988. Pergamon.
- WRIGLEY R. C. and QUAIDE W. L. (1970). Al^{26} and Na^{22} in lunar surface materials: Implications for depth distribution studies. *Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta Suppl.* 1, Vol. 2, pp. 1751-1757. Pergamon.

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Primordial Radioelements and Cosmogenic Radionuclides in Lunar Samples from Apollo 15

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Primordial Radioelements and Cosmogenic Radionuclides in Lunar Samples from Apollo 15

Abstract. Two basalts, two breccias, and two soils from Apollo 15 were analyzed by nondestructive gamma-ray spectrometry. The concentrations of potassium, thorium, and uranium in the basalts were similar to those in the Apollo 12 basalts, but the potassium:uranium ratios were somewhat higher. Primordial radioelements were enriched in the soils and breccia, consistent with a two-component mixture of mare basalt and up to 20 percent foreign component (KREEP). The abundance patterns for cosmogenic radionuclides implied surface sampling for all specimens. The galactic cosmic-ray production rate of vanadium-48 was determined as 57 ± 11 disintegrations per minute per kilogram of iron. Cobalt-56 concentrations were used to estimate the intensity of the solar flare of 25 January 1971.

The Apollo 15 mission has presented a unique opportunity to study the geochemistry of lunar surface materials by nondestructive gamma-ray spectrometry. The area explored was large and geologically diverse, and the improved sample documentation was supported by live television coverage. In addition, the

Apollo 15 manned lunar landing, unlike previous missions, was not closely preceded by an intense solar flare. Thus, the Apollo 15 material could be used in a study of the galactic cosmic-ray production rates of some short-lived radionuclides that previously were produced chiefly by solar cosmic rays. Finally,

since no quarantine requirements were imposed on samples from Apollo 15, the preliminary examination at the Lunar Receiving Laboratory (LRL) was supplemented by measurements at other laboratories.

The first Apollo 15 samples for radioactivity determination (15016,0 and 15101,1) were received from the LRL on 11 August 1971, about 9 days after lift-off from the moon. Two weeks later another pair of samples was received (15495,0 and 15601,2). Additional samples (15285,0 and 15455,0) gave valuable data on long-lived radionuclides, but because of the long decay interval before measurement they yielded little information concerning short-lived species.

The equipment and techniques of nondestructive gamma-ray spectrometry are essentially those we developed for use during the Apollo 11 and Apollo 12 missions at the LRL (1-3). Spectrum libraries for all the samples except 15455,0 were obtained from replicas that accurately reproduced the electronic and bulk densities of the lunar samples. The radioactive concentrations of the major components are given in Table 1; a more detailed analysis for components with lower concentrations is in progress. The bulk densities shown in Table 1 were obtained from the known sample weights and the measured volumes of the replicas.

The samples used in this study are representative of the varied geology associated with the Apollo 15 landing site. The locations at which they were found are shown in Fig. 1, which is a simplified map of Hadley Base drawn from the preliminary field geology reports (4). Excellent documentation exists on these samples. For the benefit of later discussion, brief sample descriptions (5) are given below.

Sample 15016,0 is a vesicular, porphyritic basalt. Its low density of 2.4 g/cm^3 suggests that it contains nearly 20 percent void space. Sample 15495,0 is a coarsely crystalline mare basalt. Its density suggests that it contains 5 to 10 percent void space due to vugs. Sample 15285,0 is a breccia of apparently homogeneous composition, with various amounts of glass coating on its surfaces. Sample 15455,0, the "black and white rock," is a dark breccia with white norite clasts. Soil sample 15101,1 from St. George Crater consists of fines of low density (about 1.2 g/cm^3) with diameters of less than 1 mm. Soil sample 15601,2 from the "terrace" of Hadley Rille consists of fines less than

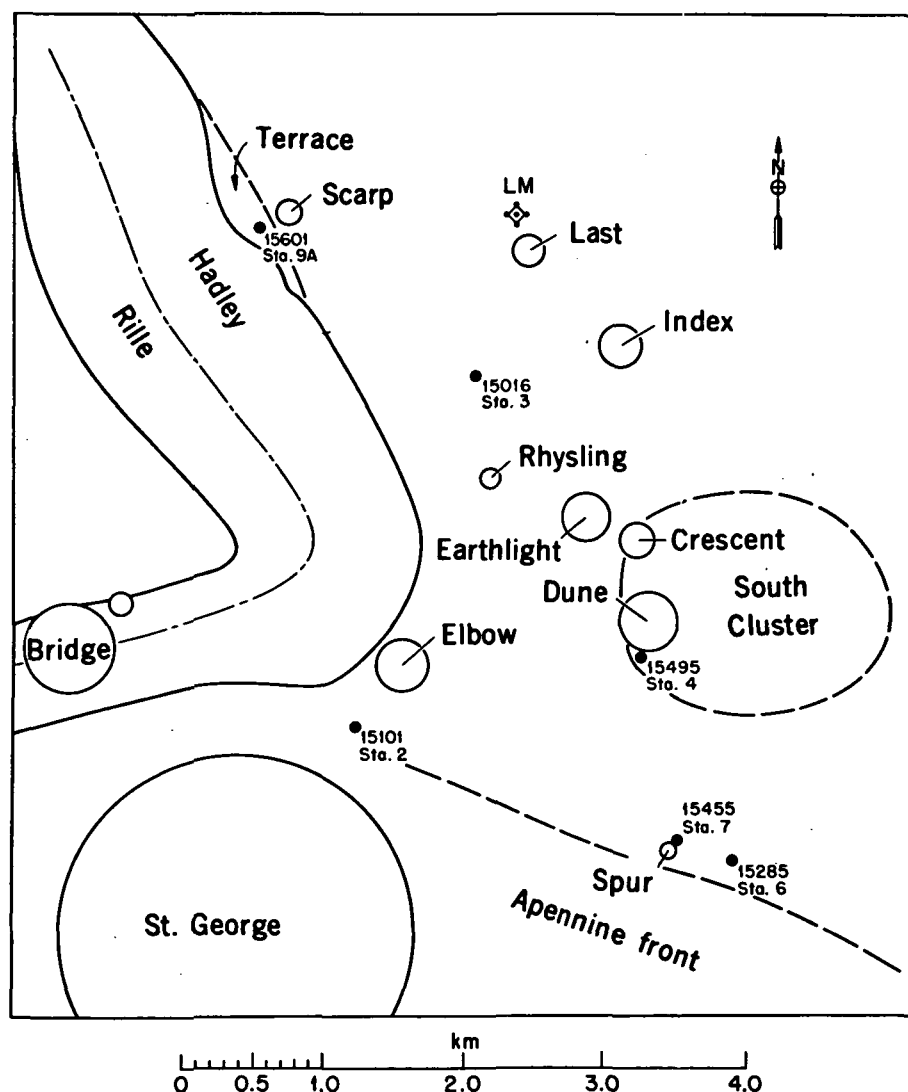


Fig. 1. Original locations of the samples described in this report. Craters are shown as open circles, and sampling stations are shown as solid black circles. The coordinates of the lunar module (LM) are $25.8^\circ \text{N } 3^\circ \text{E}$.

Table 1. Concentrations of primordial radioelements and cosmogenic radionuclides in Apollo 15 lunar samples. The values for the short-lived nuclides have been corrected for decay to 1711 hours G.M.T., 2 August 1971.

Measurement	Sample number					
	15016,0	15495,0	15285,0	15455,0	15101,1	15601,2
Petrologic type	Basalt	Basalt	Breccia	Breccia*	Fines	Fines
Weight (g)	923.7	909.0	251.3	832.8	115.6	204.3
Bulk density (g/cm ³)	2.4	2.9	2.4		~ 1.2	~ 1.7
K (ppm)†	374 ± 20	495 ± 25	1610 ± 80	900 ± 150	1484 ± 74	900 ± 45
Th (ppm)†	0.52 ± 0.02	0.60 ± 0.03	3.4 ± 0.1	2.0 ± 0.3	3.1 ± 0.3	1.8 ± 0.2
U (ppm)†	0.15 ± 0.01	0.16 ± 0.01	0.93 ± 0.05	0.53 ± 0.08	0.86 ± 0.08	0.51 ± 0.05
²⁶ Al (dpm/kg)	82 ± 4	69 ± 3	85 ± 4		120 ± 12	112 ± 11
²² Na (dpm/kg)	29 ± 2	29 ± 3	50 ± 4		44 ± 5	55 ± 6
⁴⁶ V (dpm/kg)	10 ± 2	Trace			9 ± 6	
⁵⁸ Co (dpm/kg)	16 ± 3	11 ± 2			11 ± 6	28 ± 9

* "Black and white" breccia; analysis given for dark portion by assay with Ge(Li) detector. † Terrestrial isotopic abundances and equilibria of the Th and U decay series were assumed in the standardization for assay of K, Th, and U.

1 mm in diameter and has the higher density (about 1.7 g/cm³) characteristic of mare soils from the Apollo 12 site.

The concentrations of the primordial radioelements K, Th, and U in the six samples are listed in Table 1. Since a large number of samples from previous Apollo missions have been studied by gamma-ray spectrometry, detailed comparisons with the results in Table 1 can be made.

The values of the mass ratio K/U for the earth and the moon fall into separate groups whose averages are characteristic of each planet (1, 2). The constancy of the ratio of K (a volatile element, depleted on the moon) to U (a refractory element) appears to be the result of early chemical fractionation not affected by later igneous processes. Values of K/U for lunar material are separable into the groups shown in Fig. 2, which is redrawn from our systematics (2) of Apollo 11 and Apollo 12 sample data, with the addition of recent results for Apollo 14 samples (6, 7). The K concentrations in Fig. 2 are not normalized to 18 percent Si. The correlation between the K and U concentrations implies that there is a similar relationship between K and Th, because values of the concentration ratio Th/U are nearly constant in lunar material (typically 3.6 to 3.8).

The two Apollo 15 basalts, 15016 and 15495, have K concentrations that are similar to those of the Apollo 11 low-K basalts and the Apollo 12 basalts. However, for the Apollo 15 basalts we have measured, the average value of K/U is about 2800, compared to about 2000 for the Apollo 11 and Apollo 12 basalts of the same concentration of K. In Fig. 2 the eucrite zone slightly overlaps the data region for 15016 and

15495. In fact, the K and U concentrations for the Nuevo Laredo eucrite (8), respectively, 390 and 0.14 parts per million (ppm), are almost identical to those of rock 15016.

As was the situation for the Apollo 12 samples (2), the soils and breccia from Apollo 15 are much higher in K, Th, and U than the basalts. Therefore, the soils and breccia are very likely mixtures of basalt and a foreign component high in these elements. If we assume a simple, two-component model for the soils and breccia with Apollo 15 mare basalt as one end member, then from Fig. 2 the most likely candidate for the second component is KREEP [a material high in potassium, rare earth elements, and phosphorus (9)] because

the other possibilities would yield higher K/U ratios than those observed in the soils and breccia. Even the presence of 30 percent anorthosite in the soil, because of the proximity to the lunar highlands (10), would not alter the conclusion that KREEP is the most likely foreign component, since anorthosites are very low in K, Th, and U and would simply act as diluents. Any lunar material rich in KREEP, such as the Apollo 14 samples (7), might also serve as the source of the foreign component.

Breccia 15285 is similar in composition to soil 15101 and is most likely a breccia of local origin produced by meteorite impact. Estimates based on a two-component mixture of mare basalt and KREEP suggest that these samples

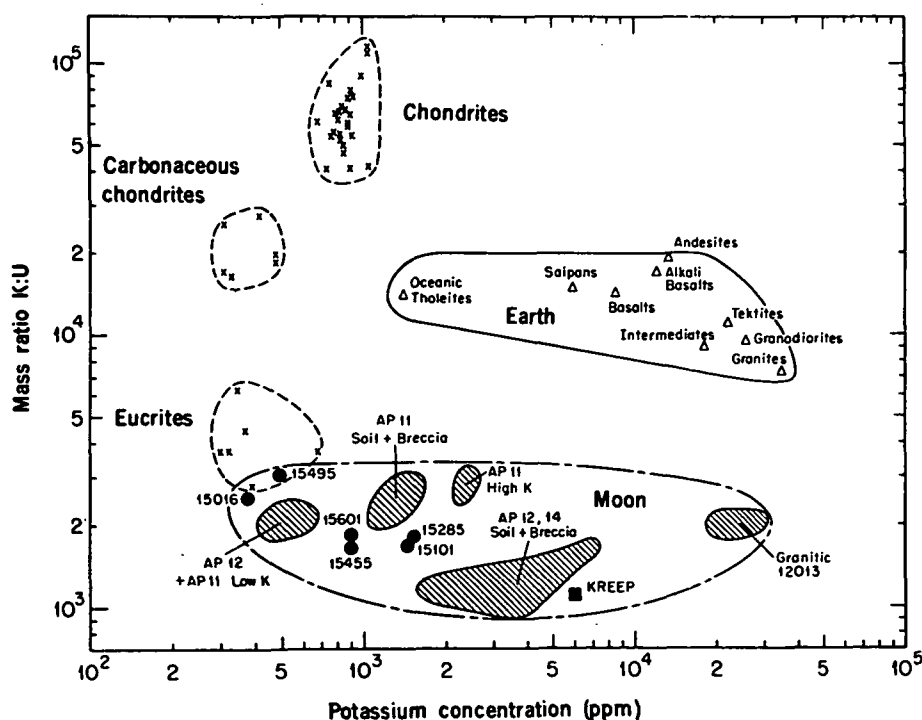


Fig. 2. Values of the mass ratio K/U as a function of the concentration of K.

contain about 20 percent KREEP. Sample 15455, the "black and white rock," is a more complex, inhomogeneous breccia. Samples 15455 and 15601 have similar concentrations of primordial radioelements and appear to be lower in KREEP (about 10 percent).

The rille soil (sample 15601) contains much less K, Th, and U than the soils and soil breccia from most of the other locations at Hadley Base (Table 1) (11). Sample 15601 was collected about 20 m from the edge of Hadley Rille. One explanation for the lower concentrations of K, Th, and U in this sample is that this area earlier was covered by a layer of lunar material as rich in KREEP as the adjoining region (up to 20 percent), but that erosion by meteorite impacts at the edge of the rille removed a large part of the layer. The effect of such impact erosion is much greater at an edge than on a horizontal plane, because the eroded material tends to fall inside the rille. It was reported that in the region under discussion the regolith was almost absent 25 m from the edge of the rille (4).

The general concentration patterns for ^{22}Na and ^{26}Al resemble those observed on previous Apollo missions. The abundance of 2.6-year ^{22}Na is a measure of the exposure to solar and galactic cosmic rays over approximately the past 10 years; the concentration of ^{26}Al is a measure of the exposure over about 2×10^6 years. Because chemical analyses are lacking for most of the samples we have examined, it is not possible to make detailed interpretations of the concentrations of ^{22}Na and ^{26}Al . However, there are no measurements that suggest strongly that the yield of ^{26}Al has not attained saturation. Further, the cosmogenic radionuclide yields, especially of ^{56}Co , suggest that the rocks were collected from the surface, in agreement with preliminary documentation.

Both soil samples also appear to have been taken from near the surface. The

high concentrations of ^{22}Na , ^{26}Al , and ^{56}Co are consistent with a sampling depth of about 3 cm for 15101 and a somewhat shallower depth of 1.5 to 3.0 cm for 15601, if it is assumed that their chemical composition is similar to that of the Apollo 12 soils.

During the preliminary examinations of the Apollo 11 and Apollo 12 samples in the LRL, we detected two relatively short-lived nuclides (1, 3); 5.7-day ^{52}Mn was determined in four rocks and 16-day ^{48}V in six rocks. The concentration of ^{48}V was well correlated with the Ti concentration of the rocks, as would be expected if most of the ^{48}V was produced by solar-flare protons through the $^{48}\text{Ti}(p,n)$ reaction. From the ^{48}V content of rock 12062, which appeared to have been buried, we inferred a yield from galactic proton bombardment of about 40 ± 20 disintegrations per minute (dpm)/kg Fe.

As shown in Table 1, we determined ^{48}V quantitatively in the first two Apollo 15 samples received, 15016 and 15101. However, the results on 15016 were superior because weak components of the gamma-ray spectra of mare basalts suffer less interference from the Th and U decay series than do the spectra of lunar soil and breccia. The concentration of ^{48}V in 15016 leads to a galactic production rate for ^{48}V of 57 ± 11 dpm/kg Fe, based on an FeO concentration (11) of 22.6 percent. This result agrees well with our earlier estimate of 40 ± 20 dpm/kg Fe, within the experimental errors, and with the value 90 ± 45 dpm/kg determined by Honda and Arnold (12) for the yield of ^{48}V in the iron meteorite Aroos.

Although no intense solar flare directly preceded the Apollo 15 mission, 77.3-day ^{56}Co was detected in some of the samples. Since ^{56}Co is almost totally produced by solar-flare protons through the $^{56}\text{Fe}(p,n)$ reaction, the ^{56}Co detected in Apollo 15 samples was produced in the solar flare of 25 January

1971. No measurements of the intensity of this flare have been published, but a comparison of the present ^{56}Co concentrations with those of the Apollo 12 samples (3) suggests that the flare of 25 January 1971 was approximately 30 percent more intense than the well-characterized event of 3 November 1969.

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References and Notes

1. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, P. R. Bell, *Science* 167, 580 (1970); in "Proceedings of the Apollo 11 Lunar Science Conference," *Geochim. Cosmochim. Acta* 2 (Suppl. 1), 1407 (1970).
2. —, in "Proceedings of the Second Lunar Science Conference," *Geochim. Cosmochim. Acta* 2 (Suppl. 2), 1159 (1971).
3. —, *ibid.*, p. 1747.
4. Apollo Lunar Geology Investigation Team, *U.S. Geol. Surv. Interagency Rep.* 32 (1971); G. A. Swann, M. H. Hait, G. G. Schaber, V. L. Freeman, G. E. Ulrich, E. W. Wolfe, V. S. Reed, R. L. Sutton, *U.S. Geol. Surv. Interagency Rep.* 36 (1971).
5. NASA Rep. MSC-03209 (1971).
6. J. S. Eldridge, G. D. O'Kelley, K. J. Northcutt, in preparation.
7. Lunar Sample Preliminary Examination Team, *Science* 173, 681 (1971).
8. G. W. Morgan and J. F. Lovering, *J. Geophys. Res.* 69, 1989 (1964).
9. C. Meyer, Jr., R. Brett, N. J. Hubbard, D. A. Morrison, D. S. McKay, F. K. Aitken, H. Takeda, E. Schonfeld, in "Proceedings of the Second Lunar Science Conference," *Geochim. Cosmochim. Acta* 1 (Suppl. 2), 393 (1971).
10. J. A. Wood, *J. Geophys. Res.* 75, 6497 (1970).
11. Lunar Sample Preliminary Examination Team, *Science* 175, 363 (1972).
12. M. Honda and J. R. Arnold, *Geochim. Cosmochim. Acta* 23, 219 (1961).
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ABUNDANCES OF PRIMORDIAL AND COSMOGENIC RADIONUCLIDES IN APOLLO 14 ROCKS AND FINES, James S. Eldridge, G. Davis O'Kelley, and K. J. Northcutt, Oak Ridge National Laboratory, P. O. Box X, Oak Ridge, Tennessee 37830

Nondestructive gamma-ray spectrometry methods developed for Apollo 11 and 12 samples (1-3) were used for the determination of K, Th, U, ^{26}Al , and ^{22}Na in eight rock samples and three soils returned by the Apollo 14 mission. All the rock samples were "fragmental," and the soils were from the "top," "middle," and "bottom" of the Soil Mechanics trench. Results of these measurements are presented in Table I.

Our suite of samples is distinguished by its uniformity in primordial radioelement content. Potassium concentrations ranged from 4000 to 5800 ppm, Th ranged from 10.9 to 15.6 ppm and U ranged from 3.1 to 4.5 ppm. The only samples from previous Apollo missions yielding this high level of primordial radioelements (average rock values) were 12013 and 12034 from the Ocean of Storms. Two soils measured from the Apollo 15 collection show K, Th, and U contents 3 to 8 times lower than the Apollo 14 soils and breccias (4).

From our simple two-component mixing predictions in Fig. 2 of reference 2, we deduce that our Apollo 14 samples contain a range of 60 - 80% foreign component (KREEP), with the average value closer to 80%. This lends support to the predictions of many investigators that the Fra Mauro formation would be rich in KREEP.

All our Apollo 14 samples fit in a tight grouping in the K/U systematics we presented in our Apollo 11 and 12 studies (Fig. 1 of reference 2). This grouping shows that the Fra Mauro samples are very similar to the dark portion of sample 12013.

Breccia 14321, the largest rock returned, originally weighed 9 kg. Our sample 14321,38 is an 1100-g piece cut from one end of the rock. Sample 14321,256 is sawdust from cutting of 14321. The good agreement for the two samples of 14321 shows that the distribution of primordial nuclides within the whole rock is uniform despite its small-scale inhomogeneity.

Cosmogenic radionuclide determinations reported in Table I show little differences from those found in previous missions with the exception of the three soil samples, which deserve special discussion. The Soil Mechanics Experiment trench was planned to be a 60-cm-deep trench at station G with one vertical sidewall to provide a means for sampling at depth. The trenching did not yield a vertical side wall; sloping occurred with walls of 60° - 80° and a maximum depth of 36 cm was achieved. Photograph AS14-64-9161 shows the degree of crumbling in the trench walls (5). Samples 14148, 14149 and 14156 shown in Table I were taken from the top, bottom, and middle, respectively of the trench and are all <1 mm sieved fractions. From the concentrations of K, Th, and U, it would appear that the soil at the trench site is uniform throughout its sampled depth of 0-36 cm. We expected to find pronounced decreases in the concentrations of the cosmogenic species ^{26}Al and ^{22}Na with depth.

Abundances of Primordial and Cosmogenic Radionuclides

James S. Eldridge

P.222

Instead, all three samples show a surprising uniformity in concentrations of these nuclides. We would predict values of ~ 40 and ~ 35 dpm/kg for ^{26}Al and ^{22}Na at the depth of 36 cm for sample 14149 (6). Due to the uniform distribution of ^{26}Al and ^{22}Na and their high concentrations at depth, we must conclude that extensive mixing occurred and sample 14149,62 is not representative of the soil at a 36-cm sampling depth. This also gives reason to question the uniformity of K, Th, and U concentrations in the different soil layers. In addition, the separation of the <1 mm fraction from the trench bottom samples has further emphasized the sampling defect since the bottom sample has a median grain size of 0.41 mm compared to 0.09 and 0.007 mm for the surface and middle trench samples (7).

Our studies with similar trench samples from Hadley Base (4) yielded the expected decrease in ^{26}Al and ^{22}Na content with increasing depth.

REFERENCES

1. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Primordial Radionuclide Abundances, Solar-Proton and Cosmic-Ray Effects, and Ages of Apollo 11 Lunar Samples by Non-Destructive Gamma Ray Spectrometry," Proceedings of the Apollo 11 Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 1, Vol. 2, pp. 1407-1423, Pergamon, 1970.
2. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Abundances of the Primordial Radionuclides K, Th, and U in Apollo 12 Lunar Samples by Nondestructive Gamma-Ray Spectrometry: Implications for Origin of Lunar Soils," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Vol. 2, pp. 1159-1168, MIT Press, 1971.
3. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Cosmogenic Radionuclide Concentrations and Exposure Ages of Lunar Samples from Apollo 12," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Vol. 2, pp. 1747-1755, MIT Press, 1971.
4. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and K. J. Northcutt, "Concentrations of Primordial Radioelements and Cosmogenic Radionuclides in Apollo 15 Samples by Nondestructive Gamma-Ray Spectrometry," this conference.
5. J. K. Mitchell, L. G. Bromwell, W. D. Carrier III, N. C. Costes, and R. F. Scott, "Soil Mechanics Experiment," in: NASA-MSC, Apollo 14 Preliminary Science Report SP-272 (1971) p. 100.
6. T. W. Armstrong, and R. G. Alsmiller, Jr., "Calculation of Cosmogenic Radionuclides in the Moon and Comparison with Apollo Measurements," Proceedings of the Second Lunar Science Conference, Geochim. Cosmochim. Acta, Vol. 2, pp. 1729-1745, MIT Press, 1971.
7. LSPET, "Preliminary Examination of Lunar Samples," in: NASA-MSC, Apollo 14 Preliminary Science Report SP-272 (1971) 117-1180.

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Abundances of Primordial and Cosmogenic Radionuclides

James S. Eldridge

P. 223

Table I. Primordial and Cosmogenic Nuclides in Apollo 14 Samples

Sample No.	Weight, g	K, ppm	Th, ppm	U, ppm	²⁶ Al, dpm/kg	²² Na, dpm/kg
<u>Clastic Rocks</u>						
169,0	78.66	5500 ± 300	14.2 ± 0.2	3.9 ± 0.1	82 ± 6	54 ± 7
170,0	26.34	5850 ± 300	14.9 ± 0.5	4.1 ± 0.1	88 ± 6	39 ± 9
265,0	65.79	4100 ± 200	10.9 ± 0.6	3.3 ± 0.2	102 ± 8	70 ± 7
271,0	96.58	5250 ± 250	15.6 ± 0.2	4.5 ± 0.3	118 ± 6	61 ± 5
272,0	46.20	4500 ± 200	11.3 ± 0.5	3.3 ± 0.2	94 ± 6	78 ± 9
273,0	22.40	4560 ± 200	11.7 ± 0.5	3.1 ± 0.2	73 ± 7	66 ± 8
321,38	1100.0	4050 ± 220	12.7 ± 0.5	3.9 ± 0.4	50 ± 20	35 ± 20
321,256	200.2	3900 ± 200	11.2 ± 0.5	3.2 ± 0.4	70 ± 7	42 ± 5
<u>Fines less than 1 mm</u>						
148,0	45.3	4150 ± 200	11.4 ± 0.5	3.3 ± 0.2	130 ± 10	74 ± 7
149,62	50.0	4650 ± 200	11.4 ± 0.5	3.2 ± 0.2	105 ± 10	66 ± 6
156,46	100.0	4410 ± 200	11.9 ± 0.5	3.3 ± 0.2	148 ± 12	68 ± 7

CONCENTRATIONS OF PRIMORDIAL RADIOELEMENTS AND COSMOGENIC RADIO-NUCLIDES IN APOLLO 15 SAMPLES BY NONDESTRUCTIVE GAMMA-RAY SPECTROMETRY,*
G. Davis O'Kelley and James S. Eldridge, Oak Ridge National Laboratory, Oak Ridge, Tenn., 37830; E. Schonfeld, NASA Manned Spacecraft Center, Houston, Texas 77058; K. J. Northcutt, Oak Ridge National Laboratory, Oak Ridge, Tenn., 37830.

Nine samples from Apollo 15 were analyzed by nondestructive gamma-ray spectrometry. The first samples (15016 and 15101) were received from the Lunar Receiving Laboratory (LRL) about nine days after liftoff from the moon. Other samples were received at intervals beginning about two weeks later.

The equipment and techniques of nondestructive gamma-ray spectrometry are essentially those we developed for use during the Apollo 11 and Apollo 12 missions at the LRL (1,2). Spectrum libraries for all samples except 15455 were obtained from replicas which accurately reproduced the electronic and bulk densities of the lunar samples. Bulk densities also were obtained from the known sample weights and measured volumes of the replicas. Results are listed in Tables 1 and 2.

As we have shown previously (1,2), mass ratios K/U for the earth and moon fall into separate groups whose average values appear characteristic of each planet. The constancy of the ratio of K (a volatile element, depleted on the moon) to U (a refractory element) appears to be the result of early chemical fractionation which is not affected by later igneous processes.

Basalts 15015, 15475, and 15495 have potassium concentrations which are similar to those of the Apollo 11 low-K basalts and the Apollo 12 basalts. However, the Apollo 15 basalts have average K/U ratios of ~ 2900 , somewhat higher than the K/U ratio of ~ 2000 for Apollo 11 and 12 basalts of the same potassium concentration. The range of K/U values for eucrites slightly overlaps the data region for the Apollo 15 crystalline rocks.

As was the situation for the Apollo 12 samples (2), the soil and breccia from Apollo 15 are much higher in K, Th, and U than the Apollo 15 basalts. Therefore, the soils and breccia are very likely mixtures of Apollo 15 basalt and a foreign component high in K, Th, and U. Our K/U systematics suggest that the most likely candidate for the foreign component is the lunar material KREEP (3). A two-component mixture requires only about 10-20% KREEP for the Apollo 15 breccia and soil samples, lower than the range of KREEP concentrations estimated for Apollo 12 samples (2). Sample 15601 and the dark portion of 15455 are similar in their primordial radioelement concentrations and are lowest in KREEP ($\sim 10\%$) of the Apollo 15 samples we have examined.

The general concentration patterns for ^{22}Na and ^{26}Al resemble those observed on previous Apollo missions. Because chemical analysis data are lacking for most of the samples in Table 2, it is not possible to make detailed interpretations of the concentrations of ^{22}Na and ^{26}Al . However, the low ratio $^{26}\text{Al}/^{22}\text{Na}$ for 15475 suggests that the ^{26}Al content did not attain

Concentrations in Apollo 15 Samples

G. Davis O'Kelley

P. 588

saturation; this implies ejection onto the lunar surface within the last 10^6 years. The cosmogenic radionuclide yields, especially of ^{56}Co , suggest that the rocks were collected from the surface, in agreement with preliminary documentation.

The radioactivity of the soil samples can be related to depth effects. The high concentrations of ^{22}Na , ^{26}Al , and ^{56}Co are consistent with a sampling depth of about 3 cm for 15101 and a somewhat shallower depth of 1.5-3.0 cm for 15601, assuming a chemical composition similar to that of Apollo 12 soils. Similarly soil 15031, taken from a 35 cm depth at the ALSEP site (Station 8), shows the low levels of cosmogenic radioactivity characteristic of deep samples. The radioactivity of 15041, taken from the top of the trench at Station 8, is consistent with sampling within the top 3 cm of the surface.

During the preliminary examination of the Apollo 11 and Apollo 12 samples in the LRL, we were able to detect in six rocks the ^{48}V produced by solar-flare protons. From the ^{48}V content of rock 12062, which appeared to have been buried, we inferred a yield from galactic proton bombardment of about 40 ± 20 dpm/kg Fe. ^{48}V

We determined ^{48}V quantitatively in the first two Apollo 15 samples received; however, the results on 15016 were superior, because weak components of the gamma-ray spectra of lunar basalts suffer less interference from the Th and U decay series than spectra of lunar soils and breccia. The concentration of ^{48}V in 15016 leads to a galactic production rate for ^{48}V of 57 ± 11 dpm/kg Fe, based on an FeO concentration (4) of 22.6%. This agrees well with our earlier estimate within the experimental errors.

Although no measurements of the intensity of the solar flare of 25 January 1971 have been published, the ^{56}Co concentrations in Table 2 suggest that this flare was slightly more intense than the well-characterized event of 3 November 1969.

* Research carried out under Union Carbide's contract with the U. S. Atomic Energy Commission through interagency agreements with the National Aeronautics and Space Administration.

1. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, Science **167**, 580 (1970); "Proc. Apollo 11 Lunar Sci. Conf.," Geochim. Cosmochim. Acta (Suppl. 1, Vol. 2) 1407 (1970).
2. G. D. O'Kelley, J. S. Eldridge, E. Schonfeld, and P. R. Bell, "Proc. Second Lunar Sci. Conf.," Geochim. Cosmochim. Acta (Suppl. 2, Vol. 2) 1159 (1971); ibid., p. 1747.
3. C. Meyer, Jr., R. Brett, N. J. Hubbard, D. A. Morrison, D. S. McKay, F. K. Aitken, H. Takeda, and E. Schonfeld, "Proc. Second Lunar Sci. Conf.," Geochim. Cosmochim. Acta (Suppl. 2, Vol. 1), 393 (1971).
4. Lunar Sample Preliminary Examination Team, "Preliminary Examination of Lunar Samples from Apollo 15," Science, in press.

Concentrations in Apollo 15 Samples

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P.589

Table 1. Concentrations of Primordial Radionuclides in Apollo 15 Samples.

Sample	Weight (g)	Density (g/cm ³)	K (ppm)	Th (ppm)	U (ppm)
<u>Crystalline Rocks</u>					
016	924	2.4	374 ± 20	0.52 ± 0.02	0.15 ± 0.01
475	288	2.9	354 ± 20	0.40 ± 0.02	0.12 ± 0.01
495	909	2.9	495 ± 25	0.60 ± 0.03	0.16 ± 0.01
<u>Breccias</u>					
285	251	2.4	1610 ± 80	3.4 ± 0.1	0.93 ± 0.05
455	833		900 ± 150	2.0 ± 0.3	0.53 ± 0.08
<u>Fines</u>					
031	100		1860 ± 95	4.3 ± 0.2	1.1 ± 0.05
041	100		1740 ± 90	4.0 ± 0.2	1.1 ± 0.05
101	116	~1.2	1484 ± 74	3.1 ± 0.3	0.86 ± 0.08
601	204	~1.7	900 ± 45	1.8 ± 0.2	0.51 ± 0.05

Table 2. Concentrations (dpm/kg) of Spallogenic Radionuclides in Apollo 15 Samples. Decays Corrected to 1711 Hours GMT, 2 August 1971.

Sample	²² Na	²⁶ Al	⁴⁶ Sc	⁴⁸ V	⁵⁴ Mn	⁵⁶ Co
<u>Crystalline Rocks</u>						
016	29 ± 2	82 ± 4	3 ± 1	10 ± 2	31 ± 4	16 ± 3
475	32 ± 3	40 ± 3	3 ± 2		23 ± 3	11 ± 5
495	29 ± 3	69 ± 3	3 ± 1	trace	25 ± 2	11 ± 2
<u>Breccia and Fines</u>						
285	50 ± 4	85 ± 4	3 ± 2		30 ± 5	
031	33 ± 3	49 ± 3			40 ± 10	
041	57 ± 4	99 ± 7	3 ± 2		33 ± 10	7 ± 5
101	44 ± 5	120 ± 12	< 4	9 ± 6	28 ± 8	11 ± 6
601	55 ± 6	112 ± 11	< 4		32 ± 8	28 ± 9